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MARTIN MARIETTA

Mercury Contamination in Poplar Creek and the Clinch River

J. W. Elwood
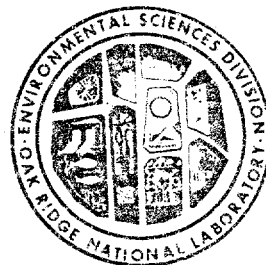
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MERCURY CONTAMINATION IN POPLAR CREEK AND THE CLINCH RIVER

J. W. Elwood

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Abstract

ELWOOD, J. W. 1977. Mercury contamination in Poplar Creek
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Laboratory, Oak Ridge, Tennessee. 70 pp.

The East Fork of Poplar Creek (EFPC), Poplar Creek (PC), and the
Clinch River (CR) below the mouth of Poplar Creek were found to be
contaminated with mercury, based on analyses and comparisons of
Hg concentrations in fish and sediments collected from these environs.
Concentrations of total mercury (Σ Hg) in muscle of all largemouth
bass collected in Poplar Creek in 1976 exceeded the FDA's proposed
action level for Hg in fish of 0.5 ppm, and 95% of the Σ Hg in these
fish was in the methyl form. The permitted level of intake by humans
of largemouth bass from Poplar Creek containing an average Σ Hg concen-
tration of 0.73 ppm, is calculated to be 40 g/day, based on the FDA
recommended maximum intake level of methylmercury. Largemouth bass
weighing more than 200 g collected from the Clinch River at the mouth
of Poplar Creek also contained Σ Hg concentrations in excess of the
FDA's proposed limit and data for this species indicate that Σ Hg levels
in the Clinch River are elevated 7 miles below the mouth of Poplar Creek
relative to concentrations in largemouth bass from Melton Hill
Reservoir. Sediment data show that Bear Creek and East Fork of
Poplar Creek are potential sources of the elevated Hg concentrations in
Poplar Creek and the Clinch River. Recommendations are given for
further defining the sources of Hg contamination in these streams and
for examining the mechanisms and rates of Hg transport in the
Poplar Creek-Clinch River system.

Introduction

In 1974, fish, benthic invertebrates, and sediments were collected from streams draining the Oak Ridge-ERDA Reservation and analyzed for several heavy metals. This sampling and analysis was conducted to provide information for the environmental impact analysis of Oak Ridge-ERDA Operations. Sediment samples collected at three locations in the East Fork of Poplar Creek (EFPC) contained approximately 25 ± 10 ppm of mercury (± 2 S.E., n = 13), while sediments in Poplar Creek (PC) below the confluence with the East Fork of Poplar Creek contained an average of 14 ± 6 ppm (n = 7) (ERDA 1975a). Two sediment samples from the Clinch River (CR) below the mouth of Poplar Creek at Clinch River Mile (CRM) 11.5 contained 17.3 and 51.9 ppm Hg. Mercury concentrations in sediments from uncontaminated control streams flowing into the East Fork of Poplar Creek contained < 0.1 ppm (ERDA 1975a). These sediment data indicated significant Hg contamination in the East Fork of Poplar Creek, Poplar Creek, and the Clinch River below the mouth of Poplar Creek. Further, muscle from several fish, particularly carp (Cyprinus carpio), collected in 1974 from the lower section of Poplar Creek in the vicinity of the Oak Ridge Gaseous Diffusion Plant (K-25) contained elevated levels of total mercury (Σ Hg) (ERDA 1975b), with concentrations in some fish exceeding the Food and Drug Administration's (FDA) proposed action level for mercury in fish of 0.5 ppm (USDHEW 1974). To verify the apparent mercury contamination in the EFPC-PC-CR drainage and to establish the extent of contamination in fish from these environs, the Environmental Sciences Division at ORNL was requested by the Oak Ridge-ERDA Operations Office to collect

fish from Poplar Creek and the Clinch River and to analyze these samples for mercury. This report contains the results of these analyses and provides comparative data for evaluating the degree of Hg contamination in these environs.

Methods

Fish were collected by electrofishing in May, June, and October 1976. All fish were weighed, measured, and frozen on dry ice. Samples were analyzed for Σ Hg by the Plant Laboratory at Y-12. Axial muscle samples of approximately 5 g were removed from each fish by cutting a rectangular section of the flesh, beginning beneath the dorsal fin and extending ventrally down the side. The skin was removed from all samples. In order to examine intra-laboratory variation in whole analysis of Σ Hg, duplicate muscle samples were removed from the first 60 fish and each duplicate was analyzed separately for Σ Hg. Duplicate samples were removed from every fifth fish thereafter if the fish was sufficiently large to take two 5-g muscle samples. Results for fish weighing < 15 g are based on whole-body analyses (i.e., entire fish, excluding the skin and gastrointestinal tract). Huckabee et al. (1974) reported no difference in Σ Hg concentration between analyses of whole fish and axial muscle in fish samples collected from uncontaminated streams in the Great Smoky Mountains National Park. This finding, however, needs statistical confirmation for fish collected from areas contaminated with mercury.

Samples of fish muscle were digested using a perchloric acid dissolution procedure (Feldman 1974), and the resulting solutions were analyzed for Σ Hg by flameless (cold-vapor) atomic absorption spectrophotometry (AAS) (Feldman 1972). To examine inter-laboratory variation in whole analysis for Σ Hg, duplicate muscle samples were removed from ten fish collected in Poplar Creek (PC-1) and were analyzed for Σ Hg at Y-12 and at the Analytical Laboratory at ORNL using flameless AAS (i.e., one duplicate from each fish was analyzed at Y-12 and the other at X-10). In addition, muscle samples from each of these ten fish were analyzed for methylmercury (MeHg) at ORNL using a gas chromatographic method with a microwave emission spectrometric detector (Talmi 1975).

To examine total procedural accuracy for Σ Hg analysis, eight samples of bovine liver reference material certified by the National Bureau of Standards were analyzed at Y-12. All concentrations of Σ Hg in fish muscle reported herein are on a part per million (ppm) fresh-weight basis.

Bottom sediments from Poplar Creek and the Clinch River were sampled for mercury analysis in July 1974 and in both July and November of 1975 and 1976 by the Environmental Management Group at the ORGDP. Sediment core samples were collected with a gravity corer from Poplar Creek Mile (PCM) 0 to PCM 5 and in the West Fork of Poplar Creek from approximately PCM 5.3 to PCM 10. Sediment samples from the Clinch River were collected upstream of the mouth of Poplar Creek from CRM 12.4 to 13.5. One sediment sample was collected below the mouth of Poplar Creek in November, 1976 at CRM 11 (Fig. 2). The upper 12 to 15 cm of

sediment from each core was sent to the Chemical Analysis Department in the Technical Services Division at ORGDP where it was wet sieved through No. 50-mesh screen (297-µ mesh opening). The < 297-µ size fraction was then dried in a steam oven at 80°C and 80 mg aliquots were analyzed for Hg using a Jarrell-Ash Emmission Mass Spectrograph (EMS) with a D.C. arc. Detection limits for this sample size was 0.01 µg Hg. The analytical precision of EMS is estimated to be no better than -50 to + 100%. Replicate analyses of sediment samples were not conducted however, so a direct estimate of analytical precision for sediment samples analyzed by this method is not available. All sediment concentrations for Hg reported herein are on a part-per-million (ppm) dry-weight basis.

Sampling Locations

Fish were collected from eight sampling areas, including the Clinch River above and below the mouth of Poplar Creek, Melton Hill Reservoir, and Poplar Creek below the confluence with the East Fork of Poplar Creek. Locations of the eight sampling areas are as follows:

1. Melton Hill Reservoir (MH).--These samples were collected in Melton Hill in the vicinity of CRM 33. As a point of reference, this sampling area is in the vicinity of the Experimental Gas-Cooled Reactor (EGCR) (Fig. 1).

2. Clinch River (CR-1).--These samples were collected in the Clinch River from the confluence with the Emory River (approximately CRM 4.5) upstream to CRM 5.5 (Fig. 2).

3. Clinch River (CR-2).--These samples were collected in the Clinch River from CRM 9.5 to CRM 10.5 (Fig. 2).
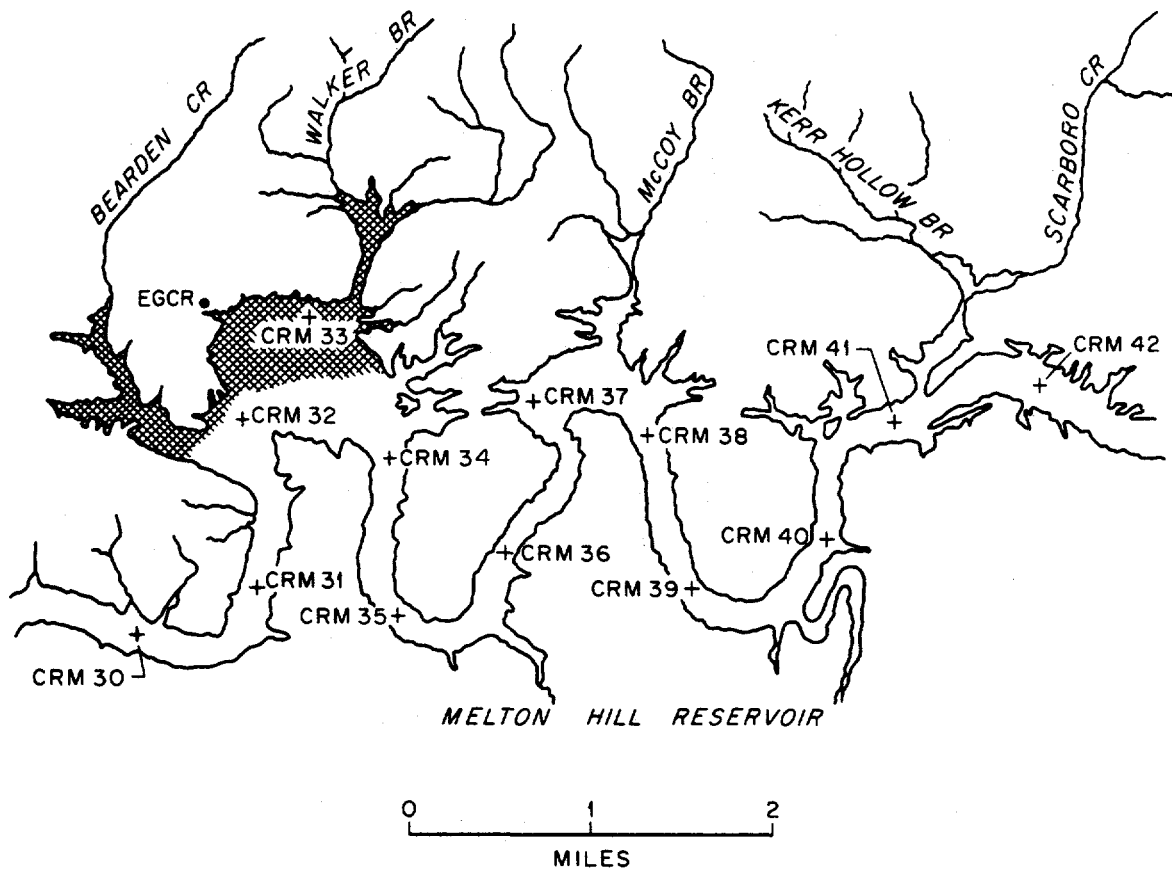
Fig. 1. Section of Melton Hill Reservoir showing the location of control sampling area MH (shaded area) near the Experimental Gas-Cooled Reactor (EGCR).
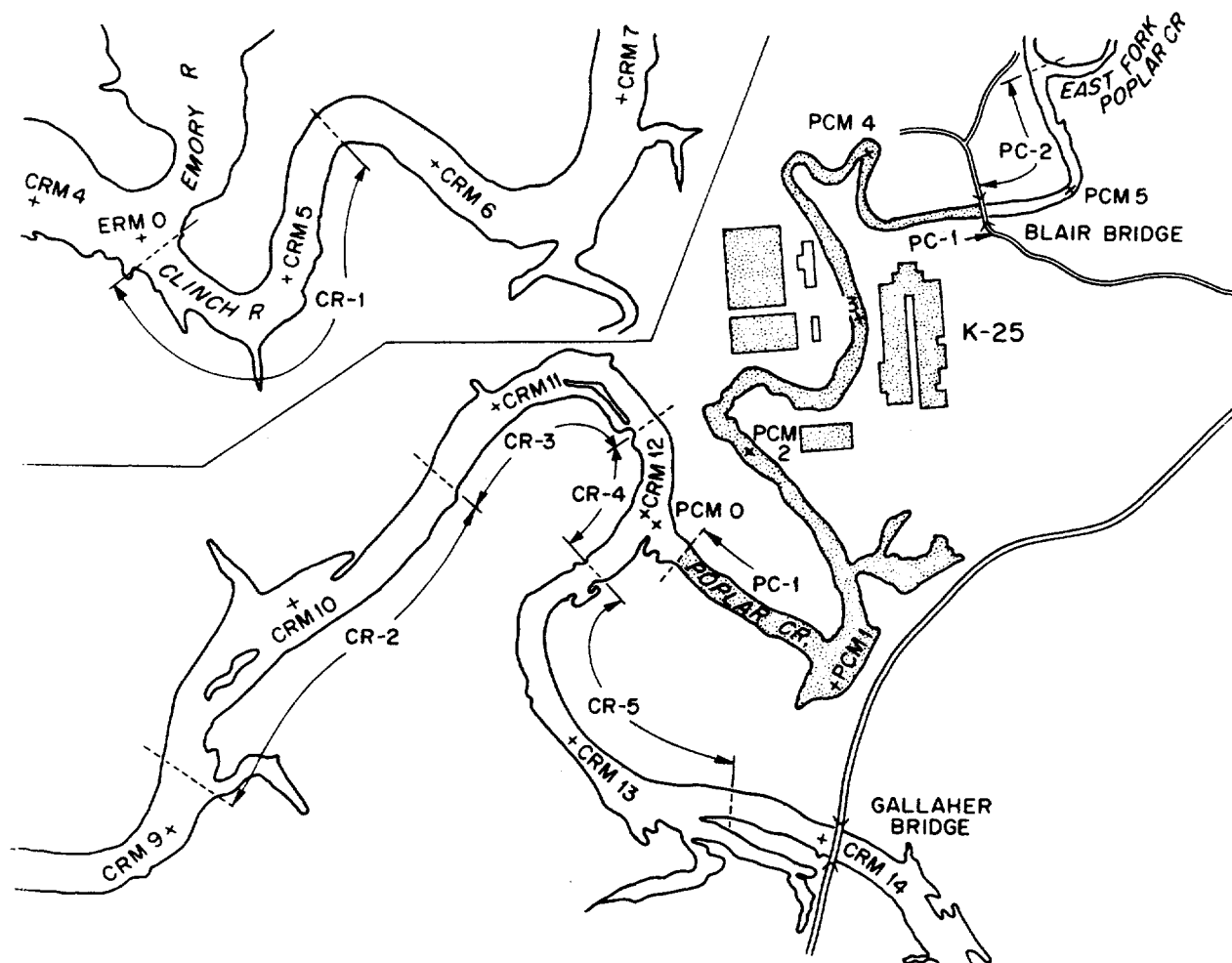
Fig. 2. Sampling locations for fish in Poplar Creek (PC) and the Clinch River (CR).

4. Clinch River (CR-3).--Samples were collected in the Clinch River from CRM 10.5 to CRM 11.5 (Fig. 2).

5. Clinch River (CR-4).--Samples were collected in the Clinch River at the mouth of Poplar Creek at CRM 12 (Fig. 2).

6. Clinch River (CR-5).--Samples were collected in the Clinch River above the mouth of Poplar Creek from CRM 12.4 to CRM 13.5 (Fig. 2).

7. Poplar Creek (PC-1).--Samples were collected in Poplar Creek from the mouth (PCM 0) upstream to Blair Bridge located at approximately PCM 4.5 (Fig. 2).

8. Poplar Creek (PC-2).--Samples were collected in Poplar Creek from Blair Bridge upstream to the confluence with the East Fork of Poplar Creek (Fig. 2).

Results and Discussion

Analytical Precision, Accuracy, and Sensitivity

Analytical precision of Σ Hg determinations in fish muscle as measured by the average coefficient of variation (C.V.) of all intra-laboratory replicate analyses (i.e., whole analysis of duplicate muscle samples removed from fish and each analyzed for Σ Hg at Y-12) was 11.2 ± 2.8% (± 2 S.E., n = 104), the C.V. ranging from 0 to 87.7%. In comparison, the average C.V. for the ten inter-laboratory replicates (i.e., one duplicate analyzed at X-10 and the other at Y-12) was 11.4 ± 4.0% (range, 2 to 18%). These results indicate both relatively good intra-laboratory precision for Σ Hg analysis in fish muscle by the Plant Laboratory at Y-12 and good intra-laboratory precision between the analytical laboratories at X-10 and Y-12.

The analytical accuracy of Σ Hg analysis given in this report is based on whole analysis of eight NBS bovine liver standards containing reference concentrations of 0.02 ppm Σ Hg. Reported concentrations of Σ Hg in all eight of the NBS standards analyzed at Y-12 was 0.02 ppm, indicating unusually high analytical accuracy. However, because the reference concentrations were known to the analytical chemist prior to the analysis for Σ Hg (R. Morrow, personal communication) and the average total-procedural precision for analyses of duplicate samples of fish muscle by the Plant Laboratory at Y-12 was ± 11%, these results probably do not provide a reliable measure of analytical accuracy for Σ Hg in samples containing unknown concentrations. The analytical sensitivity for Σ Hg in fish muscle using flameless AAS was 0.01 ppm.

Mercury Concentrations in Bluegill and Largemouth Bass

The relationship between Σ Hg concentration in axial muscle and fish size was analyzed by species for each of the eight sampling areas. Linear, semi-log and log-log regressions of Σ Hg concentration in muscle on fish weight were calculated by species for each sampling location. Based on an analysis of these regressions, it was determined that the log-log transformations generally produced the best linear fits to the data (i.e., resulted in the highest correlation coefficients between Σ Hg level and fish weight). Therefore, all regression analyses of the concentration-size relationships were done using log-log transformations. When less than five individuals of a species were collected in a sampling area the concentration-size relationship was not examined and the mean Σ Hg concentration of these individuals

was calculated. Bluegill (<u>Lepomis macrochirus</u>) and largemouth bass
(<u>Micropterus salmoides</u>) were the only species collected in sufficient
number (i.e., $n \geq 5$) to analyze the concentration-size relationship
for most sampling areas.

Statistically significant log-log regression (slope > 0 for P
< 0.10) described by the relationship $\ln Y = b_0 + b_1 \ln X$, where Y is
Σ Hg concentration in axial muscle in ppm (fresh weight), b_0 is the
intercept of the regression, b_1 is the slope of the regression, and X
is fish weight in grams, were found for bluegill from sampling areas
MH, PC-1, PC-2, CR-1, and CR-2 (Figs. 3,4,5, and 6). Regressions for
bluegill collected at PC-1 and PC-2 were not significantly different
from each other (P > 0.10), so data from these two sampling areas were
combined (Fig. 4). Slopes of regressions for bluegill collected in
the other areas were not significantly different from zero (P > 0.10),
i.e., Σ Hg concentration was independent of fish size over the range of
sizes analyzed.

Significant log-log regression of Σ Hg concentration on weight of
largemouth bass (slope > 0 for P < 0.10) were found at CR-2, CR-3, CR-4,
and CR-5 (Figs. 7, 8, 9, and 10). Slopes of regressions for largemouth
bass from the other sampling areas were not significantly different
from zero (P > 0.10). Only one largemouth bass was collected at PC-2
and, because its Σ Hg concentration was similar to that of comparable
sized largemouth bass collected at PC-1, data for this species at PC-1
and PC-2 were combined. Results for bluegill and largemouth bass
collected in the two sampling areas of Poplar Creek (PC-1 and PC-2) are
referred to hereafter as PC-C (Poplar Creek-combined).

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MHR – BG
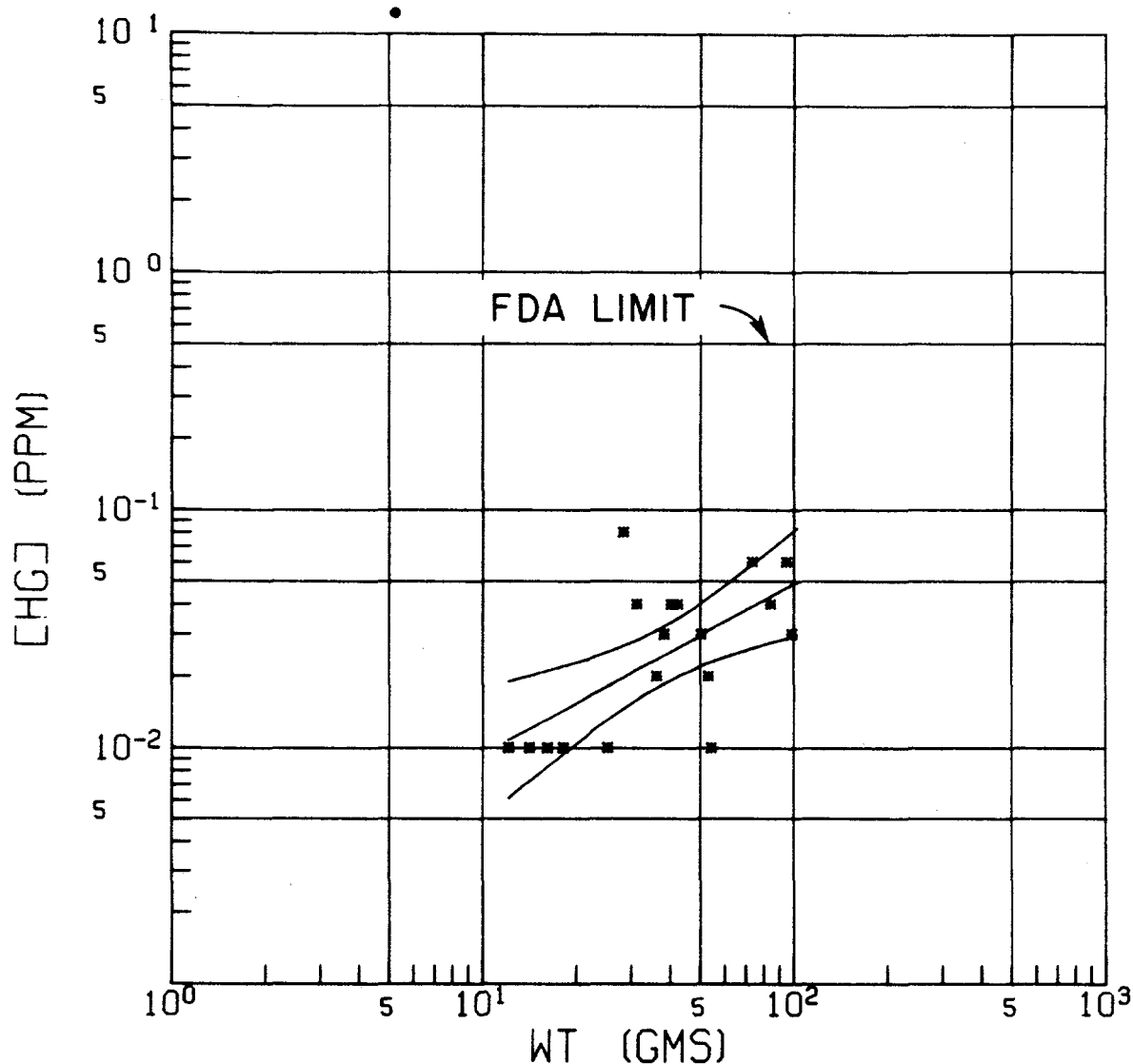
Fig. 3. Concentration-body weight relationship for Σ Hg in axial muscle of bluegill in Melton Hill Reservoir (sampling area MH). ln Y = -6.30 ($\pm$ 0.81) + 0.71 ($\pm$ 0.22) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

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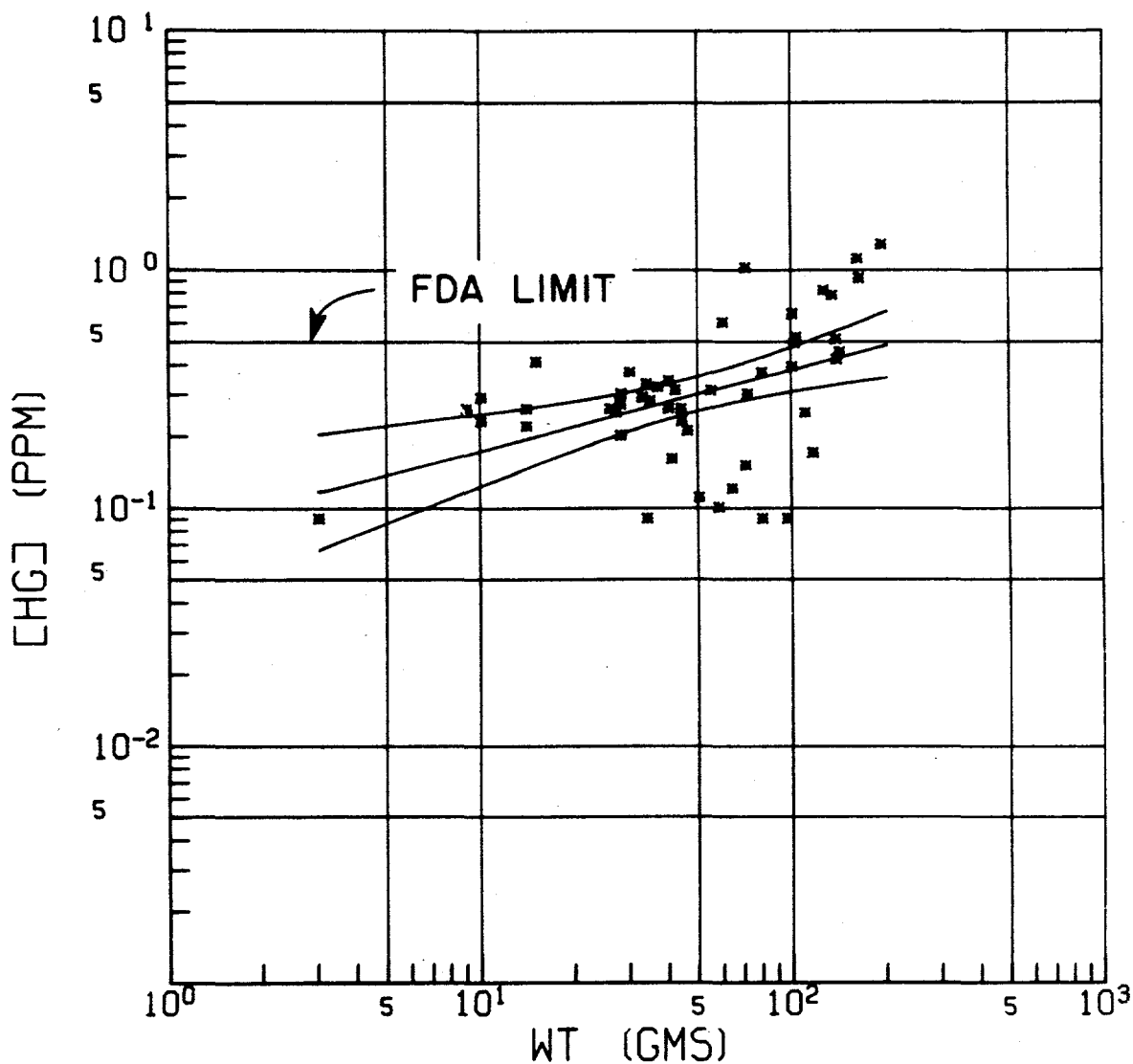
Fig. 4. Concentration-body weight relationship for Σ Hg in axial muscle of bluegill in Poplar Creek (stations PC-1 and PC-2 combined). ln Y = 2.53 ($\pm$ 0.39) + ($\pm$0.10) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

ORNL-DWG 76-19681

CR-1 BG
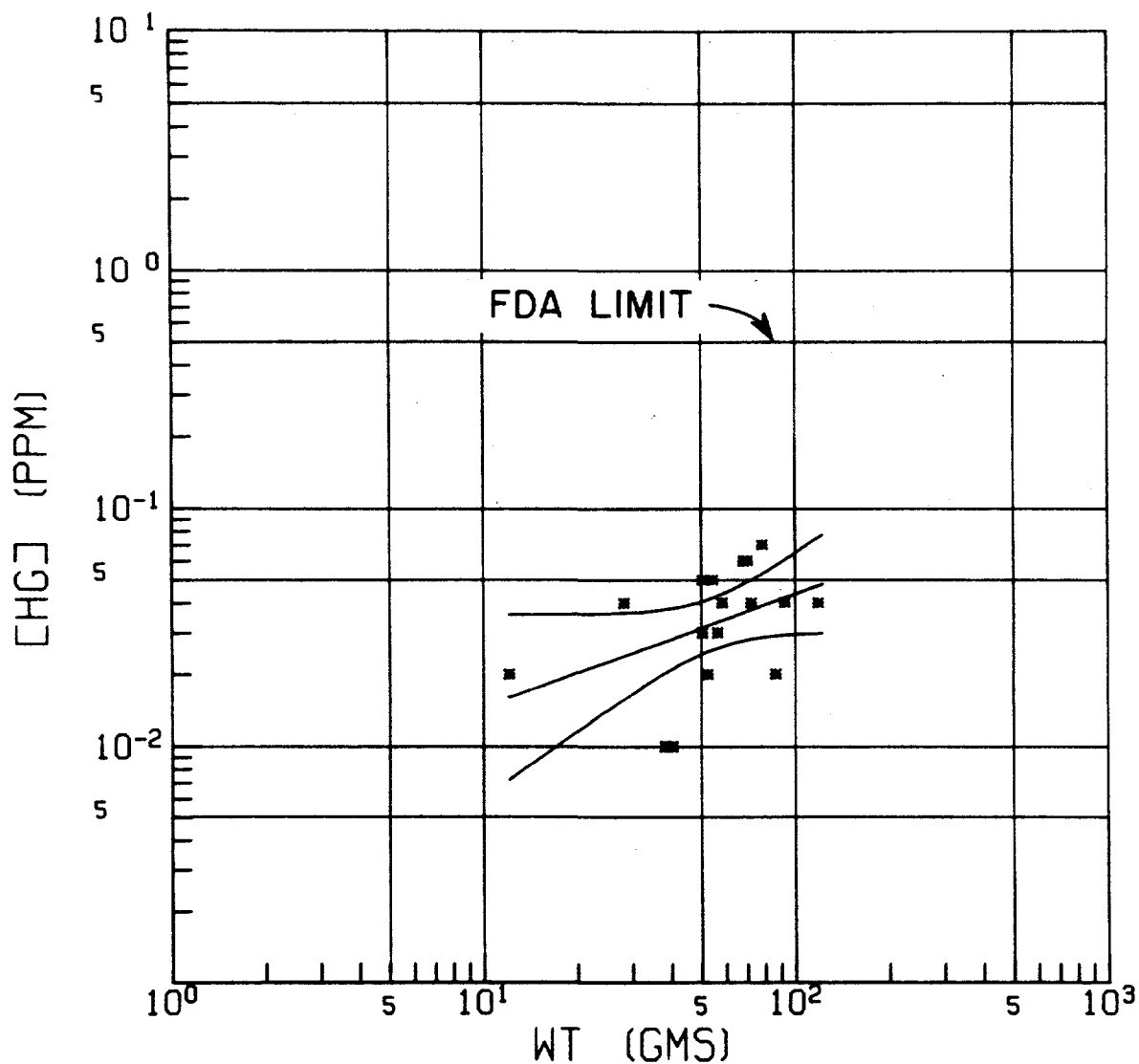
Fig. 5. Concentration-body weight relationship for Σ Hg in axial muscle of bluegill in the Clinch River at CR-1. ln Y = -5.31 ($\pm$ 1.02) + 0.47 ($\pm$ 0.25) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

ORNL–DWG 76–19682

CR–2 BG
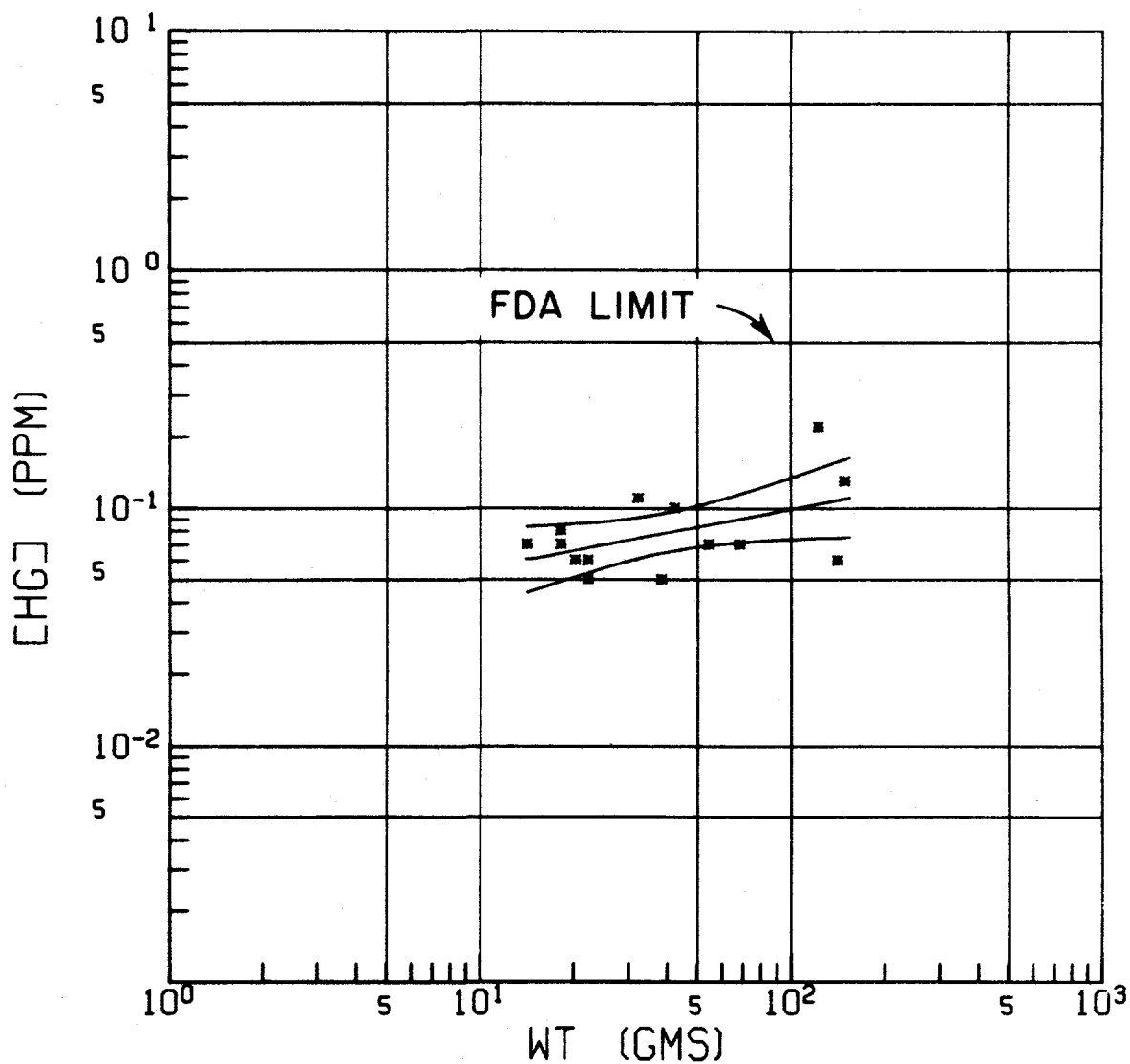
Fig. 6. Concentration-body weight relationship for Σ Hg in axial muscle of bluegill in the Clinch River at CR-2. ln Y = -3.47 ($\pm$ 0.47) + 0.25 ($\pm$ 0.13) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.
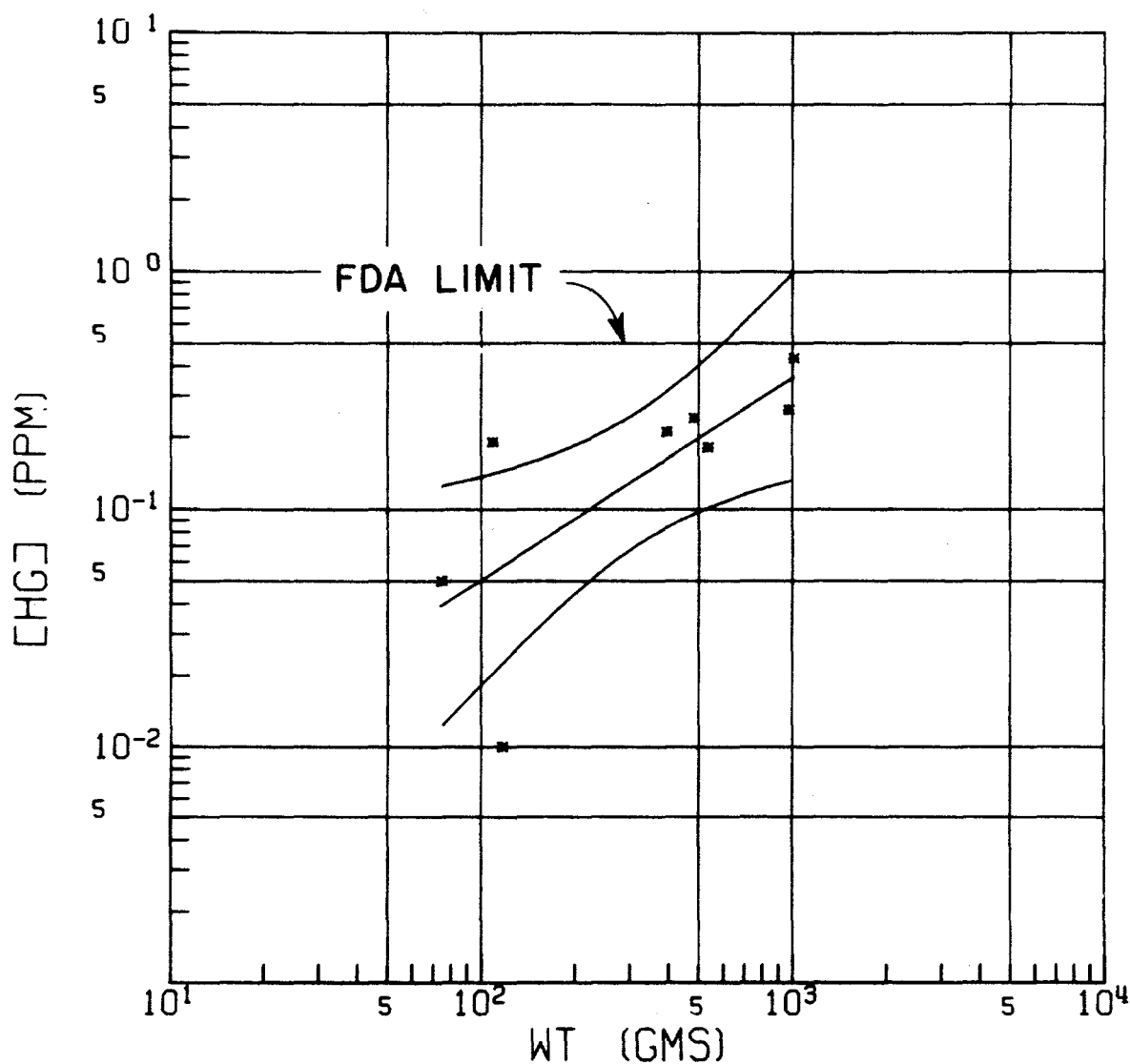
Fig. 7. Concentration-body weight relationship for Σ Hg in axial muscle of largemouth bass in the Clinch River at CR-2. ln Y = -6.90 (± 1.95) + (± 0.34) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

ORNL-DWG 76-19676

CR-3 LMB
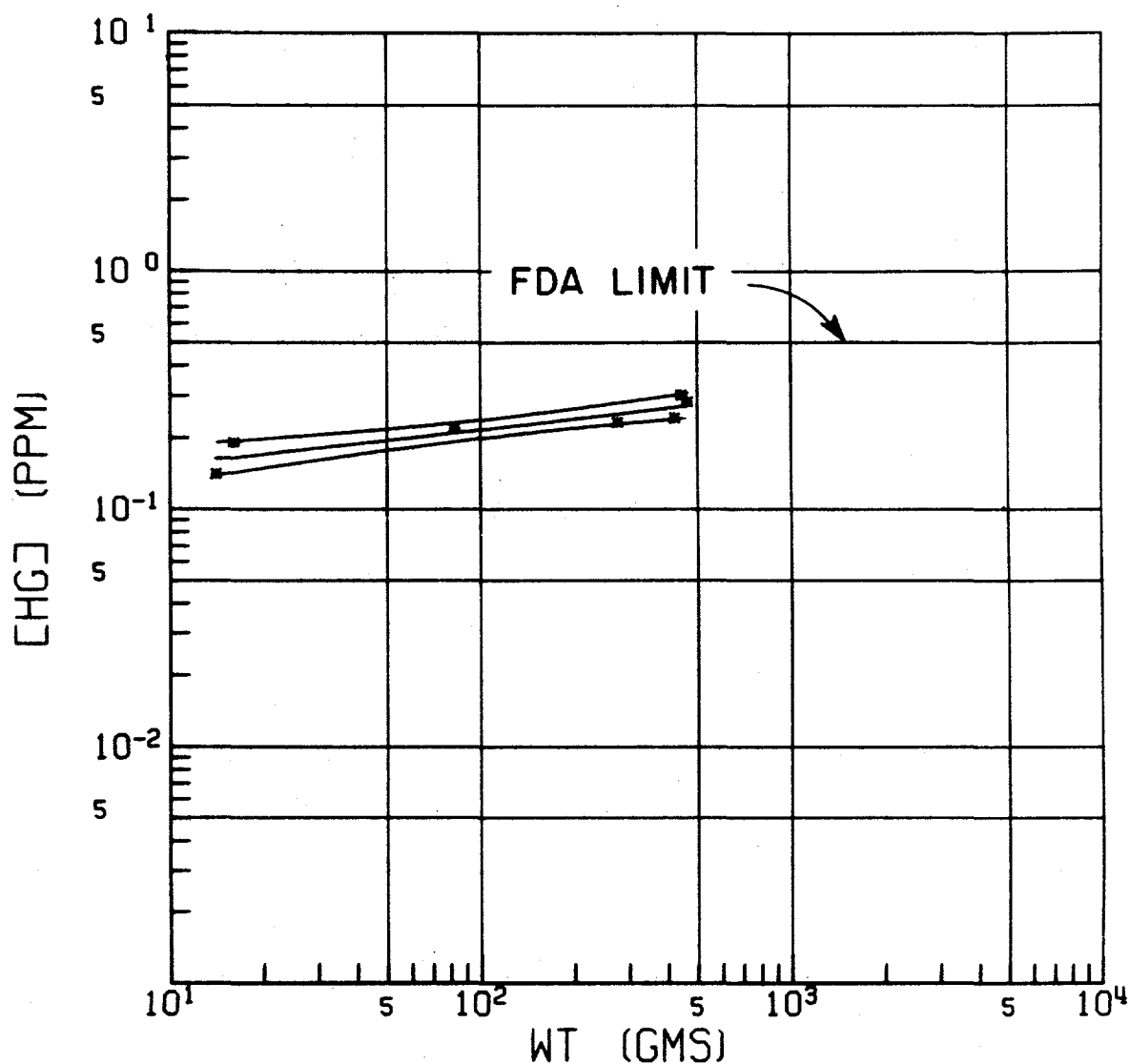
Fig. 8. Concentration-body weight relationship for Σ Hg in axial muscle of largemouth bass in the Clinch River at CR-3. ln Y = -2.21 ($\pm$ 0.16) + 0.15 ($\pm$ 0.03) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.
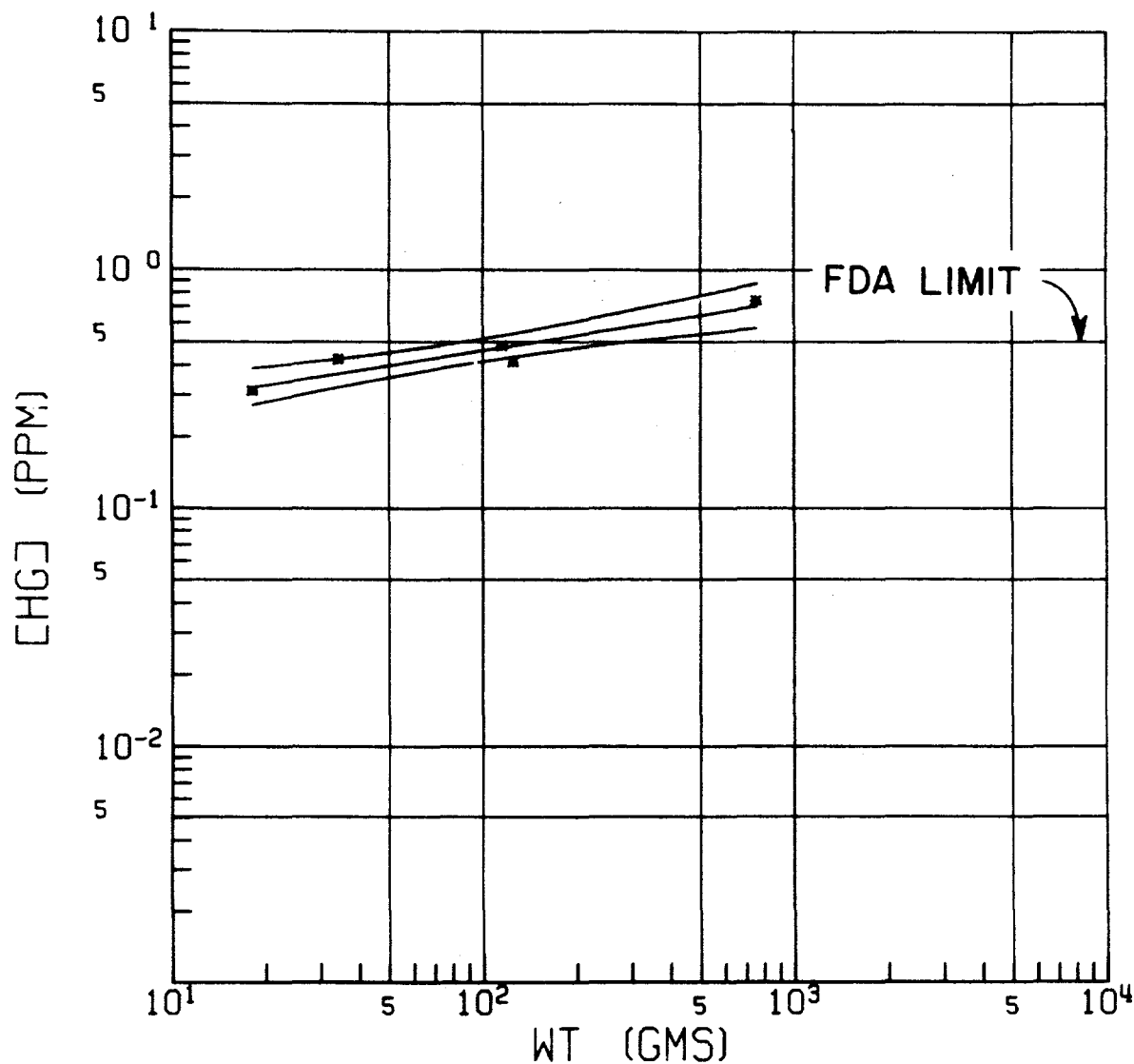
Fig. 9. Concentration-body weight relationship for Σ Hg in axial muscle of largemouth bass in the Clinch River at CR-4. ln Y = -1.73 (± 0.21) + 0.21 (± 0.04) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

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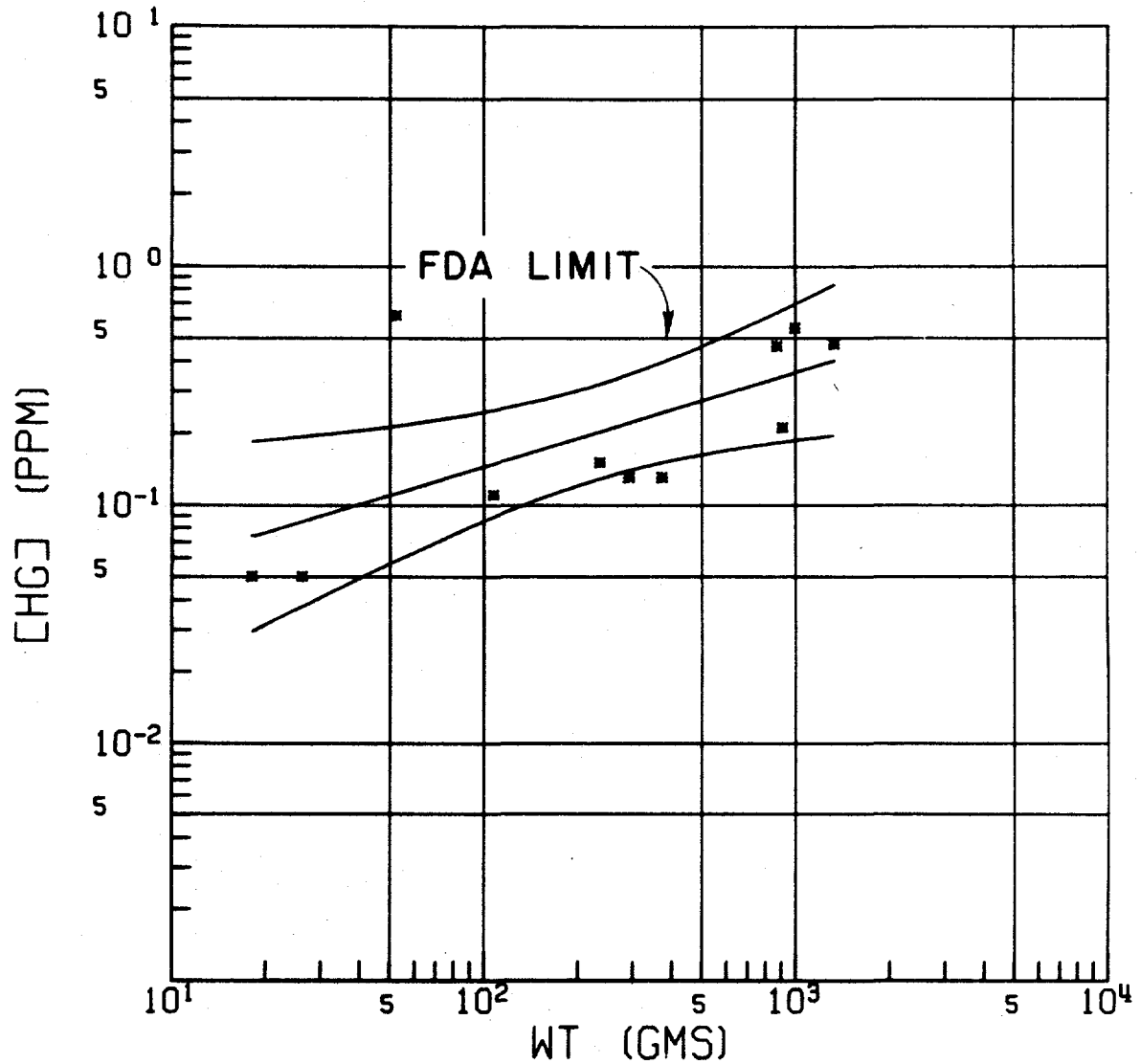
Fig. 10. Concentration-body weight relationship for Σ Hg in axial muscle of largemouth bass in the Clinch River at CR-5. ln Y = -3.75 ($\pm$ 0.93) + 0.39 ($\pm$ 0.17) ln X. Values in parentheses are standard errors of the calculated regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

Although the Σ Hg concentration body weight regressions for largemouth bass and bluegill were not statistically significant for all sampling locations, all slopes were positive except for bluegill at CR-5. These results thus tend to support the generally observed relationship of increasing Σ Hg concentration with increasing fish size (e.g., Bache et al. 1971, Olsson 1976, Anderson and Smith 1977). The fact that the regressions for some species in certain sampling locations was not statistically significant may be due to one or more factors which could obscure the Σ Hg concentration-body weight relationship. Olsson (1976), for example, found significant differences in the Σ Hg concentration-size relationship between male and female northern pike (males had higher Σ Hg concentrations than females) from a contaminated lake in central Sweden. In this study, sex of the fish was not determined so samples of each species were not separated into classes according to sex. The effect of combining data for males and females thus may have been to obscure some significant concentration-size relationships. Other possible reasons for the lack of significant correlations in some sampling areas include variation in mercury concentrations in other components of the environment (sediments, food, water) from which fish accumulate some fraction of their body burden, movement of fish into and out of areas contaminated with mercury such as Poplar Creek and the mouth of Poplar Creek, and an inability to descriminate (analytically) concentration differences at low ($\leq$ 0.01 ppm) Hg levels.

To compare levels of Hg in each species by sampling area, Σ Hg concentrations in a 275-g largemouth bass (approximate total length of 27 cm) and a 120-g bluegill (approximate total length of

18 cm) were calculated using the log-log regressions in Figs. 3 through 10. Although the fish sizes selected for comparison are arbitrary, they are typical-sized largemouth bass and bluegill which would be saved by sport fishermen for consumption. For those sampling locations in which the concentration-size relationship was not statistically significant (i.e., six out of a total of 15 regressions tested), the mean concentration of all bass and/or bluegill collected in the respective areas was calculated for comparison (i.e., Σ Hg concentration in fish muscle was assumed to be independent of fish weight in these sampling areas).

Comparisons of estimated Σ Hg levels in muscle of 275-g largemouth bass and 120-g bluegill by sampling area are shown in Figs. 11 and 12, respectively. Estimated concentrations in both species are elevated in Poplar Creek and in the Clinch River relative to concentrations in the same species in Melton Hill Reservoir (MH). All largemouth bass collected in PC-C exceeded the FDA's proposed action level for Hg in fish[1] (see footnote on pg. 55) with an average measured concentration of 0.73 ± 0.14 ppm (± 2 S.E.) and a range of 0.50 to 1.17 ppm. In comparison, the average Σ Hg concentration in largemouth bass from MH was 0.02 ± 0.01 ppm and ranged from < 0.01 to 0.05 ppm. Results for largemouth bass indicate that Σ Hg concentrations are elevated 7 miles below the mouth of Poplar Creek relative to concentrations in the same species collected in Melton Hill Reservoir (Fig. 11). Concentrations of Σ Hg in largemouth bass weighing more than 200 g (total length of 25 cm) collected at CR-4 also exceed the FDA's proposed action level for Σ Hg in fish (Fig. 9).
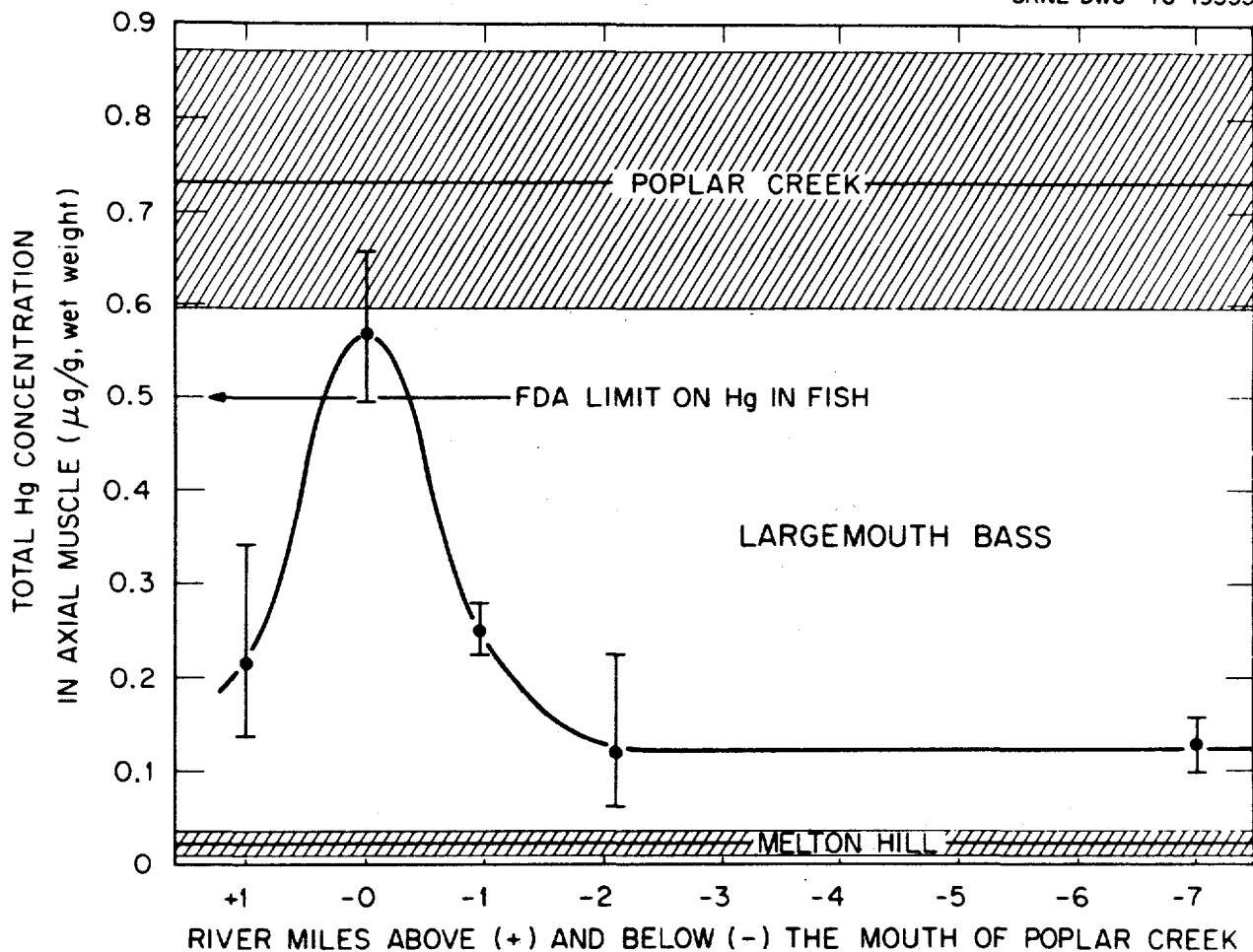
Fig. 11. Comparison of estimated mean Σ Hg concentrations in axial muscle of 275-g largemouth bass (± approximate 95% confidence limits on the calculated means) at various locations in the Clinch River above and below the mouth of Poplar Creek. Estimated mean concentrations (± 95% C.L.) in largemouth bass from Melton Hill Reservoir, and Poplar Creek are also shown for comparison.

While bluegill collected in PC-C and in the Clinch River at CR-2, CR-3, CR-4, and CR-5 contain elevated concentrations of Σ Hg relative to bluegill in MH (Fig. 12), only some of the larger (> 50 g) bluegill collected in PC-C exceed the FDA's proposed limit of 0.5 ppm (Fig. 4). The higher concentrations of Σ Hg in largemouth bass compared to bluegill, particularly at PC-C and CR-4, may be due to size or age differences between individuals of these two species, differences in rates at which mercury is absorbed and/or excreted, or to differences in Σ Hg concentrations sources from which these two species acquire their body burden of mercury. To normalize for age as a variable, Σ Hg concentration in 2-year-old largemouth bass and bluegill from each of the seven sampling areas was calculated (Table 1). The average weight of a 2-year-old largemouth bass and bluegill was estimated to be 210 and 118 g, respectively, based on reported age-size relationships for these two species from reservoirs in eastern Tennessee (Kolehmainen and Nelson 1969, Chance et al. 1975).

Results of the between-species comparison of fish of the same age show that in MH, 2-year-old bluegill are estimated to contain significantly greater (P < 0.10) concentrations of Σ Hg in axial muscle than are largemouth bass (Table 1). With the exception of CR-2 and CR-5, however, estimated Σ Hg concentrations in 2-year-old largemouth bass were significantly greater (P < 0.10) than in blue-gill of the same age in the other four sampling areas. Average calculated concentrations in 2-year-old largemouth bass at CR-5 were greater than bluegill by a factor of 2, but the difference was not statistically significant (P > 0.10). The significantly greater concentration of Σ Hg in bluegill relative to largemouth bass at MH

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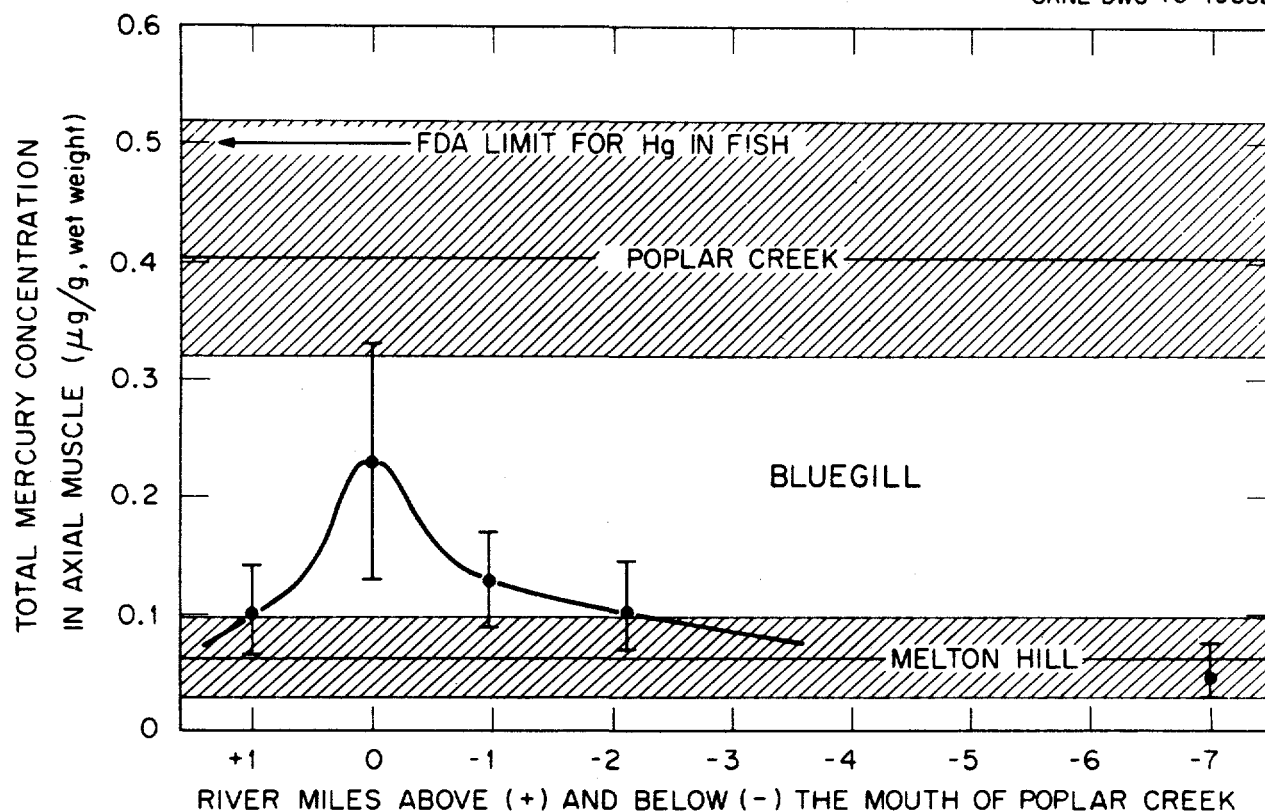
Fig. 12. Comparison of estimated mean Σ Hg concentrations in axial muscle of 120-g bluegill (± approximate 95% confidence limits on the calculated means) at various locations in the Clinch River above and below the mouth of Poplar Creek. Estimated mean concentrations (± 95% C.L.) in bluegill from Melton Hill Reservoir, and Poplar Creek are also shown for comparison.

Table 1. Test of differences in calculated mean concentrations of Σ Hg in axial muscle of a 2-year-old largemouth bass (average weight, 210 g and bluegill (average weight, 118 g) in the seven sampling areas. Values in parentheses are approximate 95% confidence limits (C.L.) on the calculated means.

| Sampling area | ppm Σ Hg (95% C.L.) | | Concentration ratio (bass:bluegill) | Significance |
	Bluegill	Largemouth bass		
MH(Melton Hill)	0.05(0.03 - 0.10)	0.02(0.01 - 0.03)	0.4	0.05[a]
CR-1(Clinch River)	0.05(0.03 - 0.08)	0.13(0.10 - 0.16)	2.6	0.05[a]
CR-2(Clinch River)	0.10(0.07 - 0.15)	0.09(0.05 - 0.19)	0.9	n.s.[b]
CR-3(Clinch River)	0.13(0.09 - 0.17)	0.24(0.22 - 0.27)	1.8	0.05[a]
CR-4(Clinch River)	0.23(0.13 - 0.33)	0.54(0.47 - 0.62)	2.4	0.05[a]
CR-5(Clinch River)	0.10(0.06 - 0.14)	0.19(0.12 - 0.31)	1.9	n.s.[b]
PC-C(Poplar Creek)	0.40(0.32 - 0.51)	0.73(0.59 - 0.87)	1.8	0.05[a]

[a]Mean concentrations in the two species are statistically different (P < 0.10).

[b]Calculated mean concentrations are not statistically different (P > 0.10).

may be due to the fact that a small size range of largemouth bass was collected in Melton Hill compared to the size range of this species collected in the other six sampling areas. Also, several of the largemouth bass and bluegill collected in MH had Σ Hg concentrations below the detection limit of 0.01 ppm. Concentration of Σ Hg in these fish, however, was assumed to be 0.01 ppm when the concentration-size relationship was examined. The effect of this assumption may have been to obscure significant Σ Hg concentration-size relationships for these two species in MH, resulting in an underestimate of the Σ Hg concentrations in 2-year-old fish collected in this sampling area.

With the exception of results at MH and CR-2, data in Table 1 indicate that concentrations of Σ Hg in axial muscle of 2-year-old largemouth bass are significantly greater than in bluegill of the same age; the average concentration ratio of largemouth bass to bluegill for CR-1, CR-3, CR-4, and PC-C is 2.2 ± 0.4. This difference in Σ Hg concentrations between the two species could be due to differences in size and/or feeding habits. To normalize for size as a variable, Σ Hg concentrations in muscle were calculated for a 118-g largemouth bass and bluegill in each of the seven sampling areas (Table 2). Ignoring the results at MH for the reasons discussed previously, data in Table 2 show that 118-g largemouth bass contain significantly greater (P < 0.05) concentrations of Σ Hg than bluegill of the same size (but which are older than 118-g largemouth bass) at four of the six sampling areas. The average concentration ratio of largemouth bass to bluegill weighing 118 g from these four areas is 2.0 ± 0.4.

Table 2. Test of differences in calculated mean concentrations of Σ Hg in axial muscle of a 118-g largemouth bass and bluegill in the seven sampling areas. Values in parentheses are the approximate 95% confidence limits (C.L.) on the calculated means.

Sampling area	ppm Σ Hg (95% C.L.)		Concentration ratio (bass:bluegill)	Significance
	Bluegill	Largemouth bass		
MH(Melton Hill)	0.05(0.03 - 0.10)	0.02(0.01 - 0.03)	0.4	0.05[a]
CR-1(Clinch River)	0.05(0.03 - 0.08)	0.13(0.10 - 0.16)	2.6	0.05[a]
CR-2(Clinch River)	0.10(0.07 - 0.15)	0.06(0.02 - 0.15)	0.6	n.s.[b]
CR-3(Clinch River)	0.13(0.09 - 0.17)	0.22(0.20 - 0.24)	1.7	0.05[a]
CR-4(Clinch River)	0.23(0.13 - 0.33)	0.48(0.42 - 0.54)	2.1	0.05[a]
CR-5(Clinch River)	0.10(0.06 - 0.14)	0.15(0.09 - 0.26)	1.5	n.s.[b]
PC-C(Poplar Creek)	0.40(0.32 - 0.51)	0.73(0.59 - 0.87)	1.8	0.05[a]

[a]Mean concentrations in the two species are significantly different (P < 0.05).

[b]Mean concentrations are not significantly different (P > 0.05).

To examine whether the significantly greater concentration of
Σ Hg in largemouth bass compared to bluegill in most of the sampling
locations was a result of a greater rate of accumulation, as estimated
by slopes of the log-log regressions, a test was made to determine if
the estimated slopes for the two species at each site differed sig-
nificantly. Results show that except for PC-C where the slope was
significantly greater for bluegill than for largemouth bass, the
estimated rates of Hg accumulation are not significantly different
between the two species (Table 3). This suggests that the difference
in Σ Hg levels between these species observed in most sampling
locations is manifested in earlier stages of the life history (e.g.,
eggs, fry, fingerlings) which were not sampled and analyzed in this
study. With the higher concentrations of Σ Hg in largemouth bass,
it is not unreasonable to expect that the ovaries in this species
would also contain proportionally higher concentrations relative
to bluegill which would be passed on to the next generation. Thus,
a concentration difference in embryos of these two species would be
established. McKim et al. (1976) found that a considerable amount
of mercury was passed from the parents to the eggs of brook trout
chronically exposed over three generations to methylmercuric chloride.
His results show that the concentration in the embryos was proportional
to the concentration of Hg in water and that the concentration differ-
ence between treatments was maintained in the eggs, embryos, and adults
through three generations. The factor(s) initially producing the
concentration differences between largemouth bass and bluegill ob-
served in this study is unknown but may be a result of differences

Table 3. Comparison of estimated slopes (± standard error) of log-log regressions of Σ Hg concentration in muscle on body weight of largemouth bass and bluegill in the Clinch River, Melton Hill Reservoir, and Poplar Creek. Values in parentheses sample sizes.

Sampling Location	Bluegill	Largemouth bass	Significance
CR-1	0.47 ± 0.25(18)	0.06 ± 0.12(11)	n.s.[a]
CR-2	0.25 ± 0.13(14)	0.85 ± 0.34(8)	n.s.[a]
CR-3	0.11 ± 0.16(20)	0.15 ± 0.03(7)	n.s.[a]
CR-4	0.46 ± 0.53(16)	0.21 ± 0.04(5)	n.s.[a]
CR-5	-0.09 ± 0.32(15)	0.39 ± 0.17(11)	n.s.[a]
MH	0.71 ± 0.22(18)	0.26 ± 0.43(11)	n.s.[a]
PC-C	0.34 ± 0.10(50)	0.03 ± 0.09(8)	0.05[b]

[a]Slopes not significantly different from each other ($P > 0.10$).

[b]Slopes significantly different ($P < 0.05$).

in exposure times to mercury in the environment. Largemouth bass
tend to be a longer-lived species than bluegill and thus are exposed
longer to their environment. The difference may also be a result of
a greater concentration in the source(s) of mercury from which large-
mouth bass accumulate their body burden. Although both species of
fish are predators, largemouth bass are primarily piscivorous (fish
eaters) whereas bluegill tend to feed predominantly on benthic
invertebrates.

Total Mercury in Other Fish Species

Several other fish species were collected in the eight sampling
areas and analyzed for Σ Hg. Results for fish collected at PC-1
and PC-2 were combined (PC-C). Sample size for individual species
in many of the sampling areas was inadequate to analyze the
concentration-size relationship. Significant log-log regressions
of Σ Hg concentration on fish weight (slope > 0 for P < 0.10) were
found, however, for suckers (Catostomus spp.) at CR-2 and CR-5
(Figs. 13 and 14) and for white crappie (Pomoxis annularis) at CR-5
(Fig. 15). Concentration-size relationships were not significantly
different from zero for carp (Cyprinus carpio) at MH, CR-5, and
PC-C, or for suckers at CR-3 and PC-C.

A comparison of Σ Hg concentration by species and sampling
area is shown in Table 4. Results for most species show the same
trend as that observed for bluegill and largemouth bass (Figs. 11
and 12), i.e., elevated concentrations in fish from PC-C and CR-4
relative to concentrations in fish collected from the other samp-
ling areas. Concentrations of Σ Hg in several fish collected in
PC-C and CR-4 exceeded the FDA's proposed limit of 0.5 ppm Σ Hg.

ORNL-DWG 76-20314

CR-2 SUCKER
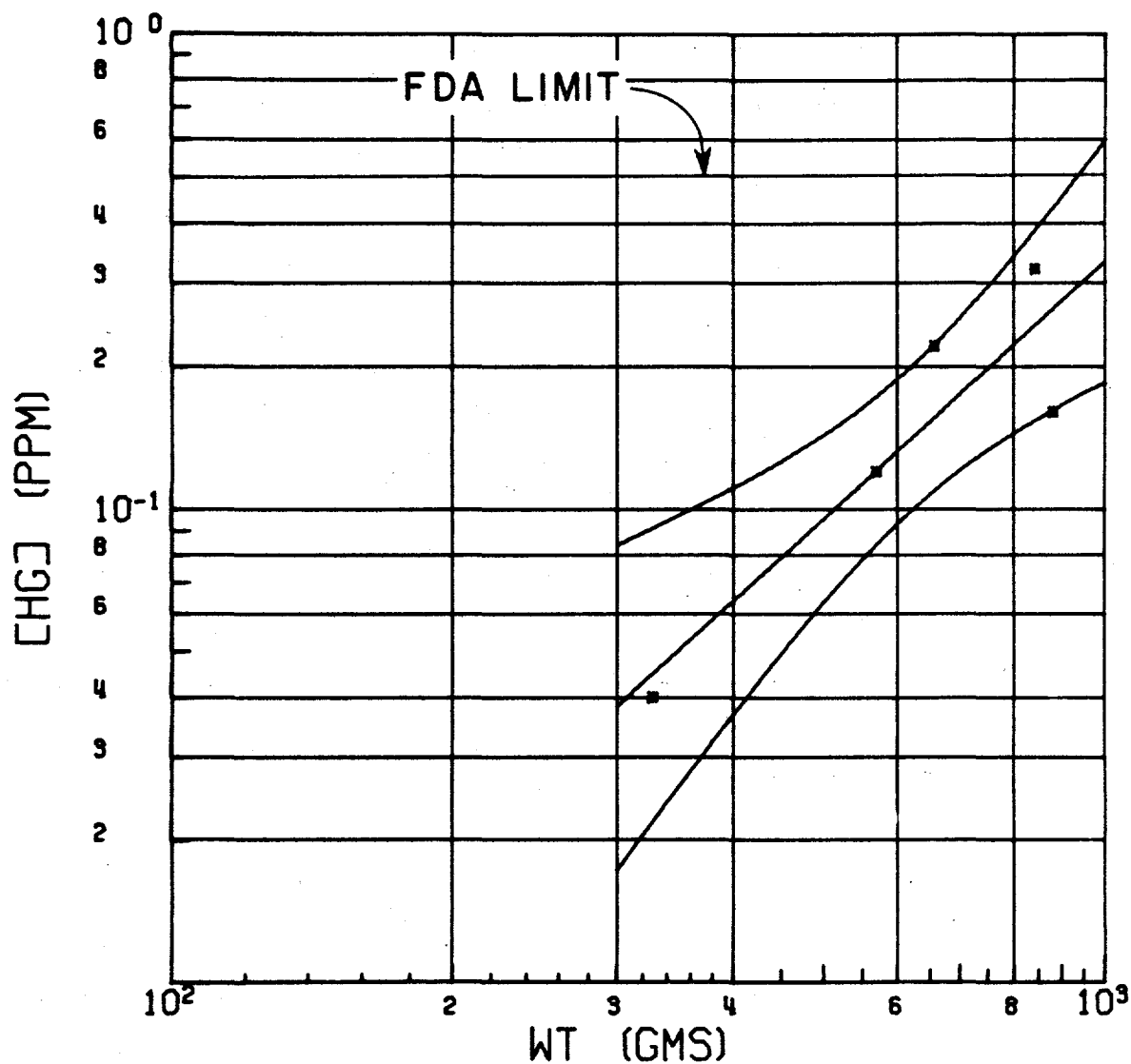
Fig. 13. Concentration-body weight relation for Σ Hg in axial muscle of suckers (*Catostomus* spp.) at CR-2. ln Y = -13.50 (± 3.15) + 1.79 (± 0.49) ln X. Values in parentheses are standard errors of the regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

ORNL-DWG 76-20313

CR-5 SUCKER
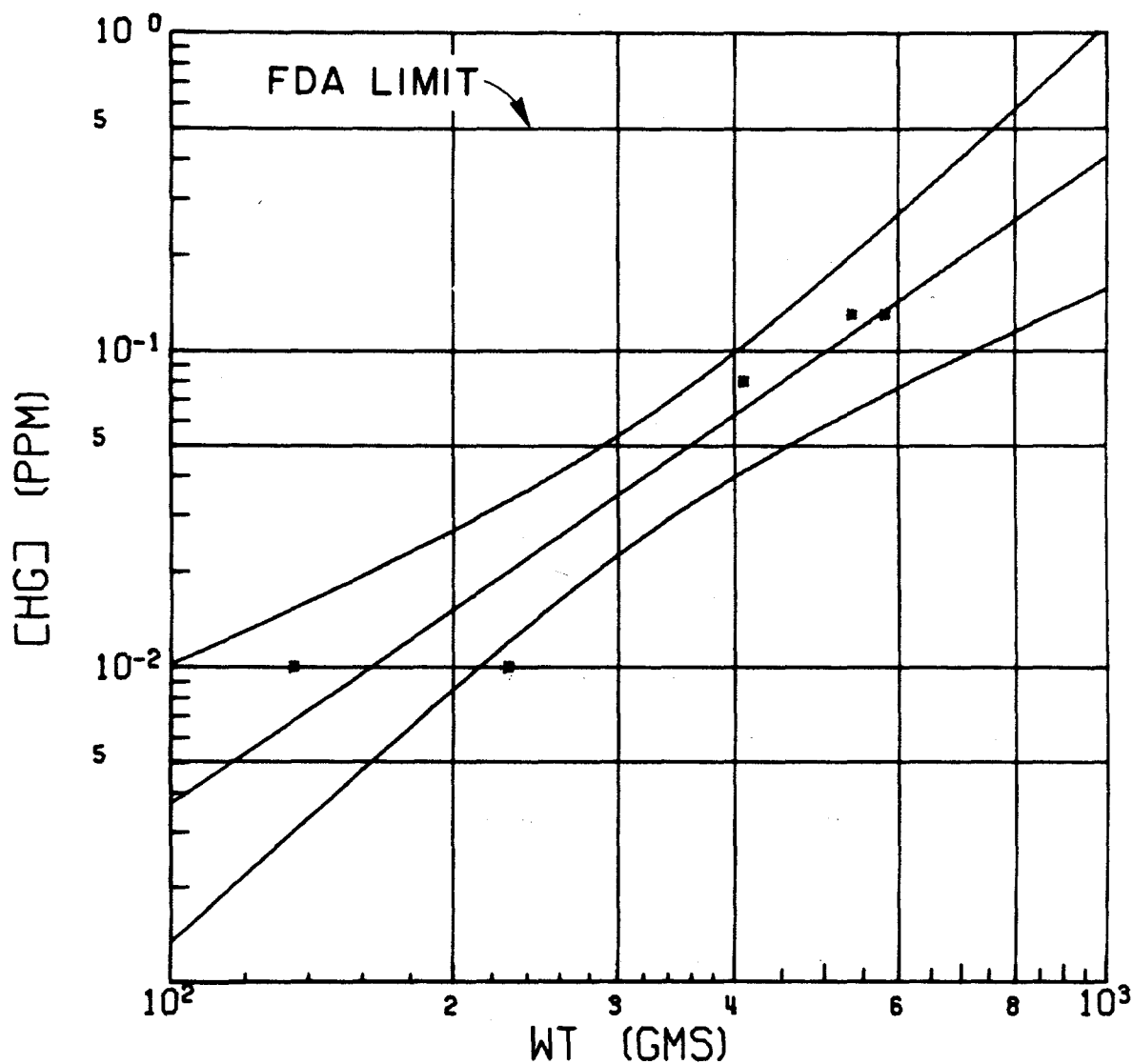
Fig. 14. Concentration-body weight relationship for Σ Hg in axial muscle of suckers (<u>Catostomus</u> spp.) at CR-5. ln Y = -15.00 ($\pm$ 2.25) + 2.04 ($\pm$ 0.39) ln $\overline{X}$. Values in parentheses are standard errors of the regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

ORNL-DWG 76-20315
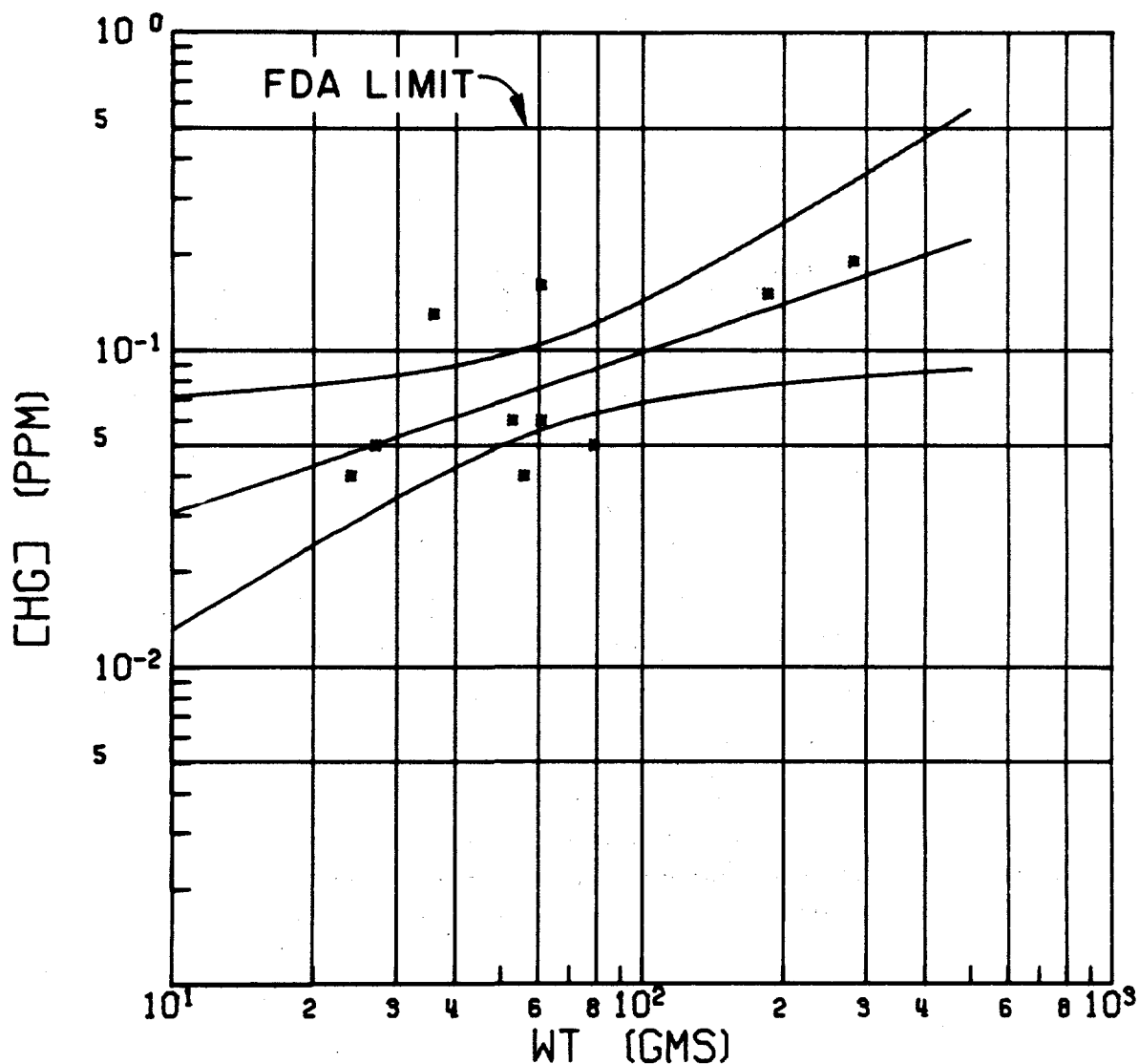
Fig. 15. Concentration-body weight relationship for Σ Hg in axial muscle of white crappie at CR-5. ln Y = -4.66 ($\pm$ 0.90) + 0.51 ($\pm$ 0.21) ln X. Values in parentheses are standard errors of the regression coefficients, upper and lower lines are the approximate 95% confidence limits on the estimated regression line.

Table 4. Mean concentrations of Σ Hg in axial muscle of fish from the Clinch River (CR), Melton Hill Reservoir (MH), and Poplar Creek (PC), ppm fresh weight. Values in parentheses are approximate 95% confidence limits (C.L.) on the calculated means.

Sampling area	Carp	Bigmouth buffalo	Suckers	White crappie
MH	0.11 ± 0.06(0.05-0.17) n = 6	--------	-------	0.03, n = 1
CR-1	0.15 ± 0.11(0.14-0.26) n = 2	--------	0.21 ± 0.23(0-0.44) n = 3	0.03, n = 1
CR-2	0.27 ± 0.12(0.15-0.39) n = 4	0.12 ± 0.08(0.04-0.20) n = 2	0.10[a](0.06-0.14)	0.05 ± 0.03(0.02-0.08) n = 2
CR-3	0.07, n = 1	0.61, n = 1	0.28 ± 0.14(0.14-0.42) n = 5	--------
CR-4	0.34 ± 0.16(0.18-0.50) n = 4	1.88 ± 0.20(1.68-2.08) n = 2	0.15 ± 0.11(0.04-0.26) n = 2	--------
CR-5	0.20 ± 0.03(0.17-0.23) n = 5	0.61, n = 1	0.10[b](0.06-0.17)	0.14[c](0.08-0.25)
PC-C	0.48 ± 0.23(0.25-0.71) n = 6	0.71 ± 0.65(0.06-1.36) n = 5	0.27 ± 0.14(0.13-0.41) n = 7	0.42 ± 0.22(0.20-0.64) n = 4

[a]Calculated concentration in a 500-g sucker based on the concentration-size relationship in Fig. 13.

[b]Calculated concentration in a 500-g sucker based on the concentration-size relationship in Fig. 14.

[c]Calculated concentration in 200-g white crappie based on the concentration-size relationship in Fig. 15.

Popular Creek is utilized by sport fishermen, particularly in the spring when spawning runs of adult white bass (<u>Roccus</u> <u>chrysops</u>) and sauger (<u>Stizostedion</u> <u>canadense</u>) migrate up Poplar Creek. It was hoped that a large sample of these two species could be collected in Poplar Creek in order to determine and compare the mercury levels in migratory species which probably remain in the area of high mercury contamination for a relatively short period of time (< one month) with the levels in resident sport species (e.g. largemouth bass, bluegill, white crappie). Unfortunately, sampling in May 1976 was initiated after the spawning runs of sauger and white bass were completed. As a result, only four sauger and two white bass were collected (see p. 34). Since some individuals of these two species may be residents in the area of the Clinch River and Poplar Creek that is contaminated with Hg, it is essential to collect the samples when the spawning runs are occurring. The small sample size of sauger and white bass collected in May 1976 is unfortunately, inadequate to draw any conclusions about the levels of Σ Hg in migratory species that move into and through the area of mercury contamination.

Methylmercury in Fish

Results of methylmercury (MeHg) analysis for ten fish collected in PC-C show that 94.8 ± 10.7% of the Σ Hg in muscle is in the methyl form. The percent MeHg in the ten fish analyzed ranged from 65 to 103%. Several published studies have reported that essentially all Hg in fish muscle is in the methyl form (<u>inter alia</u>, Westoo 1969, Zitko et al. 1971, Hildebrand et al. 1976, Bishop and Neary 1974).

Concentrations of Σ Hg in axial muscle of white bass and sauger collected from Poplar Creek (PC-C) and the Clinch River (CR-2) in May 1976.

Site	Species	Weight (g)	Σ Hg Concentration ($\mu g/g$, fresh weight)
PC-C	Sauger	219	0.30
PC-C	Sauger	835	1.35
PC-C	White bass	328	0.20
PC-C	White bass	666	0.18
CR-2	Sauger	120	0.08
CR-2	Sauger	450	0.12

Bishop and Neary (1974) reported an average MeHg content of 88.9 ± 7% of total in 16 species of freshwater fish, with a range of 82 to 95%. They also reported that the proportion of MeHg was independent of fish size, length, or weight and that there were no significant differences in the percent MeHg between species.

While all forms of mercury are toxic, methylmercury (MeHg) is more toxic chronically than the other forms. Assuming fish contain 0.5 ppm Hg and all the mercury is in the methyl form, the FDA's proposed action level permits a fish consumption rate for humans of 60 g/day (i.e., daily consumption of 60 g of fish containing 0.5 ppm MeHg) (USDHEW 1974). The permitted level of intake of largemouth bass from Poplar Creek (PC-C) which contain an average Σ Hg concentration of 0.73 ppm would be 40 g/day.[2]

Comparative Data on Mercury in Freshwater Fish

Concentrations of mercury occurring naturally in tissues of fish removed from any known source of mercury contamination have not been well defined; published values range from 0.02 to 0.2 ppm (Harriss 1971, Wallace et al. 1971, Huckabee 1974). Huckabee (1974) reported average Σ Hg concentrations in rainbow trout (Salmo gairdneri) and brook trout (Salvelinus fontinalis) from streams in the Great Smoky Mountains National Park of 0.036 and 0.018 ppm, respectively. Comparative data on Σ Hg in other species from several reservoirs in the southeastern United States are shown in Table 5. In general, reported data for Melton Hill Reservoir seem to support the findings obtained in this study for MH, with Σ Hg concentrations in bluegill and largemouth bass of < 0.1 ppm. Likewise, reported concentrations in bluegill and largemouth bass from Pickwick Reservoir (0.40 and 0.64 ppm, respectively), which received significant inputs of mercury from a chloralkali prior to 1971,[3] are comparable to average values in the same species from Poplar Creek (0.37 and 0.73 ppm). These results suggest that in aquatic environments which receive little, if any, anthropogenic inputs of mercury, concentrations in fish muscle are < 0.1 ppm, while in contaminated environments, such as Pickwick and Cherokee Reservoirs and Poplar Creek, Σ Hg concentrations consistently exceed 0.1 ppm.

Mercury in Sediments

Concentrations of Σ Hg in surface sediments collected from Poplar Creek and the Clinch River from 1974-1976 are given in Table 6. There is considerable variability (< 0.1 to 300 ppm Σ Hg) in the concentration

Table 5. Mean concentrations (± 2 S.E.) of Σ Hg in fish from reservoirs in the southeastern United States

Site	Species	Σ Hg concentration (ppm, fresh weight)
Norris Reservoir	All species	< 0.05[a]
Melton Hill (CRM-43)	Bluegill	< 0.2[a]
	Largemouth bass	< 0.2[a]
	Buffalo	< 0.2[a]
Watts Bar (TRM 155)[b]	Bluegill	< 0.05[a]
	Largemouth bass	< 0.05[a]
	White crappie	< 0.05[a]
Watts Bar (TRM 579)[b]	Carp	< 0.05[a]
	White crappie	< 0.05[a]
Fort Loudon (TRM 618 and 634)[b]	Bluegill	< 0.05[a]
	Largemouth bass	< 0.05[a]
Fontana Reservoir	Bluegill	< 0.05[a]
	Largemouth bass	< 0.05[a]
Pickwick	Bluegill	0.40 ± 0.07[a]
	Largemouth bass	0.64 ± 0.07[a]
	Carp	0.58 ± 0.04[a]
	Hog sucker	0.53 ± 0.52[a]
	Spotted sucker	0.23 ± 0.07[a]
	Sauger	0.74 ± 0.17[a]
Cherokee: 118[c]	Bluegill	0.32 ± 0.07[d]
148[c]	Bluegill	0.13 ± 0.03[d]
172[c]	Bluegill	0.15 ± 0.04[d]
118[c]	Largemouth bass	0.34 ± 0.16[d]
148[c]	Largemouth bass	0.11 ± 0.04[d]
172[c]	Largemouth bass	0.19 ± 0.04[d]

[a]Source: Tennessee Valley Authority (TVA). 1972. Mercury concentrations in fish flesh, May 1970-May 1972--Survey of TVA Reservoirs. Division of Environmental Research and Development, Water Quality Branch. (unpublished report). 34 pp.

[b]TRM = Tennessee River Mile.

[c]River miles below chlor-alkali plant on North Fork Holston River at Saltville, Virginia.

[d]Source: Oak Ridge National Laboratory (ORNL). 1976. Ecology and Analysis of Trace Contaminants, Progress Report, October 1974-December 1975. ORNL/NSF-22, Oak Ridge, Tennessee. 200 pp.

Table 6. Mean concentrations of Σ Hg in the < 297 μ size fraction of surface sediments collected in Poplar Creek above and below the confluence with EFPC and in the Clinch River. Unless indicated otherwise, sample size (n) includes samples with concentrations < detection limit of 0.1 ppm.

Site	July, 1974		July, 1975		November, 1975		July, 1976		November, 1976	
PCM 0-1	20	(n=1)	8.6(< 0.1-20)	(n=23)	5.7(2-10)	n=3	2.0	n=3	8.0(2-20)	n=3
PCM 1-2	11.5(3-20)	(n=2)	45.0(< 0.1-300)	n=15[a]	7.0(4-10)	n=2	7.5(5-10)	n=2	6.0(2-10)	n=2
PCM 2-3	7.5(5-10)	(n=2)	8.9(< 0.1-30)	n=23	1.2(< 0.1-2)	n=4	7.8(1-10)	n=4	45.2(0.5-125)	n=3
PCM 3-4	———————		4.6(< 0.1-20)	n=14	———————		———————		———————	
PCM 4-5.2	20 (10-30)	(n=2)	3.4(< 0.1-10)	n=20	65.5(< 0.1-250)	n=8	3.3(1-10)	n=9	21.2(< 0.1-100)	n=8
PCM 6-10	———————		all < 0.1	n=4	———————		———————		———————	
$\bar{X}$ for PCM 0-5.2	14.0(3-30)	n=7[c]	14.2(< 0.1-300)	n=72[b,c]	26.9(< 0.1-250)	n=11[c]	4.6(1-10)	n=18[c]	21.3(< 0.1-125)	n=14[c]
CRM 12.3-13.5	< 0.1	(n=1)	all < 0.1	n=5	< 0.1	n=1	< 0.1	n=1	0.1	n=1
CRM 11	———————		———————		———————		———————		20	n=1

[a]The calculated mean concentration for this section when the two samples containing 300 ppm are omitted is 5.8 ppm.

[b]The calculated mean concentration for PCM 0-5.2 when the two samples containing 300 ppm are omitted is 6.3 ppm.

[c]n includes only those samples with concentrations > detection limits.

of Σ Hg sediments collected from PCM 0 to PCM 5. Average Σ Hg concentration for all sediment samples collected in this section of Poplar Creek was 14.2 ± 48.3 (n = 74). The maximum concentration of 300 ppm was found in two samples collected between PCM 1 and PCM 2 in July 1975. These two samples have a significant effect on the calculated average concentration of Σ Hg in Poplar Creek sediments as indicated by the fact that the mean concentration and standard deviation are 6.3 ± 5.6 ppm, respectively, when these values are omitted.[4]

The reason for the large variation in Hg concentrations in this section of Poplar Creek both within one sampling area and over time is unknown but probably is due in part to the analytical method used for analyzing the sediment samples. Emission mass spectrometry (EMS) is an inherently imprecise method for mercury analysis. The precision is probably reduced even more for solid sediment samples because of the difficulty in obtaining a homogeneous sample matrix for analysis with a relatively small quantity of sediment sample ($\sim$ 80 mg) (i.e., textural differences in samples submitted for Σ Hg analysis). Several studies have shown that mercury in river and lake sediments is associated predominantly with the smallest particle sizes which have the highest specific surface areas (i.e., cm^2/g sediment) and also frequently the highest content of organic matter (Cranston and Buckley 1972, Thomas 1972, Thomas and Jaquet 1976). Cranston and Buckley (1972), for example, found that 99% of the mercury in sediments from the La Have River, Nova Scotia, was associated with sediment particles < 60 μ in size. Coarser particles, which include the sand-sized particles (62 - 2000 μ), are usually inert with respect

to mercury, and these can be considered diluents in sediment samples (Thomas 1972). In comparative studies of mercury in sediments, it is thus necessary to assure that samples are texturally similar or that the particle-size distribution of samples is known. Sediment samples from Poplar Creek consisted of particles < 297 μ and, thus, may have contained a substantial amount of larger sediment particles (fine to medium sand) which would be expected to contain little or no mercury.

Concentrations of Σ Hg in all sediment samples collected in the West Fork of Poplar Creek (PCM 10) were < 0.1 ppm, which would seem to eliminate this stream as a source of the elevated levels of Σ Hg in sediments and fish in the lower section of Poplar Creek (PCM 0 to PCM 5) and in the Clinch River below the mouth of Poplar Creek.

Sediment data for other tributaries in the Poplar Creek Drainage are limited. Only one sediment sample was collected in the East Fork of Poplar Creek (EFPC) in 1975, and it contained < 0.1 ppm of Hg. Sediments collected from EFPC in previous years, however, show elevated levels of mercury. In 1970, for example, mercury concentrations in sediments collected at three locations on EFPC (200 yards below New Hope Pond, in the area of Wiltshire Estate, and in the area of the Oak Ridge Country Club) ranged from 0.9 to 11.3 ppm, while sediments from New Hope Pond which drains into Poplar Creek contained 63 ppm of Hg (Sanders 1970). The analytical method and size fraction analyzed was not specified. In 1974, Reece reported concentrations of Σ Hg in bulk surface

sediments analyzed with AAS from five locations in EFPC, ranging

from 3 to 18 ppm (dry weight) (Reece 1974).

Reported concentrations of Σ Hg in sediments from Bear Creek

are also highly variable. With the exception of a single sample con-

taining 6 ppm, Σ Hg concentrations in bulk sediments, analyzed by AAS,

collected from Bear Creek at five locations in 1974 were < 1 ppm,

while sediments from Grassy Creek, an uncontaminated control stream

located near Bear Creek, contained < 0.05 ppm Σ Hg (Reece 1974).

Concentrations of Σ Hg in sediments collected from Bear Creek

in conjunction with another environmental sampling program (ERDA

1975a), however, ranged from < 0.2 to 152 ppm with a mean of

13.4 ± 41.7 ppm (n = 13). Part of the variation in measured

Σ Hg in sediments from Bear Creek and the East Fork of Poplar Creek

is due to the fact that bulk sediments were analyzed so all samples

were probably not of similar texture. There may also be substantial

spatial and temporal variation in Σ Hg associated with sediments in

these streams as a result of variation in discharge. In any case,

results for Bear Creek and the East Fork of Poplar Creek suggest that

both of these streams may contribute to the elevated levels of Σ Hg

in sediments and fish in Poplar Creek and the Clinch River.

The low (< 0.1 ppm) concentrations of Σ Hg in sediment samples

collected in the Clinch River above the mouth of Poplar Creek indicate

that the Poplar Creek drainage is the major contributor to the ele-

vated levels of Hg in sediments and fish below the mouth of Popular

Creek. Additional sampling and analysis of the < 53 μ sediment frac-

tion in the Clinch River above the mouth of Popular Creek should

confirm this.

Comparison of Mercury Concentrations in Sediments from Freshwater Environments

The average concentration of Σ Hg in the continental crust is estimated to be 0.08 ppm based on the assumption of equal abundance of basaltic and granitic rocks exposed to weathering in the continental crust (Taylor 1964), with reported concentrations in sedimentary rocks higher than in igneous rocks by a factor of 2 to 3 (Rankama and Sahama 1950). Sediments downstream of ore deposits or contaminated industrial discharges may contain from 0.2 to 200 ppm Hg (USGS 1970). Reported concentrations of Σ Hg in sediments of most lakes, rivers, and reservoirs, however, average < 1 ppm (Table 7), although these data must be interpreted with caution because of textural differences in the sediment samples. Sediments from freshwater environments which are known to have received significant anthropogenic inputs of Hg, such as Pickwick Reservoir, North Fork of the Holston River below Saltville, Virginia, and the western basin of Lake Erie associated with the Detroit River, contain Σ Hg concentrations that exceed 1 ppm (Table 7). Comparison of data in Table 7 with sediment data from Poplar Creek (Table 6), and the East Fork of Poplar Creek indicates that a significant amount of mercury contamination has occurred in this drainage.

Sources of Mercury in the PC-CR Drainage

Whether the elevated concentrations of Hg in sediments are due to recent releases from Oak Ridge-ERDA facilities at Y-12 and ORGDP or are a result of previous releases which have been retained in sediments of this drainage is unknown.

Table 7. Mean mercury concentrations (± 1 S.D.) in surface sediments of freshwater environments, including southeastern reservoirs. Values in parentheses are ranges.

Site	Sediment fraction analyzed	Σ Hg concentration ($\mu g/g$, dry weight)		
Lake Superior	Bulk sediment	0.083 ±	0.056	(0.004- 0.584)[a]
Lake Michigan (southern part)	Bulk sediment	0.146 ±		(0.030- 0.380)[a]
Lake Erie	Bulk sediment	0.582 ±	0.555	(0.008- 2.929)[a]
Lake Ontario	Bulk sediment	0.651 ±	0.507	(0.032- 2.100)[a]
Average for Great Lakes	Bulk sediment	0.347 ±		(0.004- 9.500)[a]
Fort Loudon (TRM[b] 604-650)	Not given	0.29 ±	0.10	(< 0.05 - 0.38)[c]
Watts Bar (TRM 555-579)	Not given	0.18 ±	0.01	(0.17 - 0.19)[c]
Chickamauga (TRM 484-508)	Not given	1.83 ±	1.91	(< 0.05 - 4.3)[c]
Nickajack (TRM 426-459)	Not given	0.37 ±	0.27	(0.21 - 0.91)[c]
Guntersville (TRM 351-424)	Not given	0.68 ±	0.61	(< 0.05 - 1.8)[c]
Pickwick (TRM 208-259)	Not given	7.82 ±	15.22	(< 0.05 - 73.0)[c]
Kentucky (TRM 24-203)	Not given	0.73 ±	0.56	(< 0.05 - 1.7)[c]
N. Fork Holston River (above Saltville, VA)	< 44μm	0.13 ±	0.02	(0.11 - 0.16)[d]
N. Fork Holston River (2 to 82 miles below chlor-alkali plant at Saltville, VA)	< 44	8.12 ±	6.32	(2.4 - 20.8)[d]
Cherokee Reservoir	< 44	1.92 ±	0.23	(0.48 - 2.13)[d]
Poplar Creek, TN (PCM[e] 0-5)	< 297	14 ±	4.06	(< 0.1 -300)[f]
West Fork of Poplar Creek, TN (PCM10)	< 297	All < 0.1[f]		

[a]Sources: Thomas, R. L., and J. M. Jaquet. 1976. Mercury in the surficial sediments of Lake Erie. J. Fish. Res. Board Can. 33:404-412; and Thomas, R. L. 1974. The distribution and transport of mercury in the sediments of the Laurentian Great Lakes system. pp. 1.1-1.16. In Proc., Int. Conf. on Transport of Persistent Chemicals in Aquatic Ecosystems, Ottawa.

[b]TRM = Tennessee River Mile.

[c]Source: Churchill, M., W. Fulkerson, P. A. Krenkel, and W. Sanders. 1971. A plan for studying mercury deactivation in Pickwick Reservoir. (unpublished report). 31 pp.

[d]Sources: Hildebrand, S. G., A. W. Andren, and J. W. Huckabee. 1976. Distribution and bioaccumulation of mercury in biotic and abiotic compartments of a contaminated river-reservoir system. pp. 211-232. In Andrew, R. W., P. V. Hodson, and D. E. Konasewich (eds.), Toxicity to Biota of Metal Forms in Natural Waters, Proc., Workshop International Joint Commission on the Great Lakes, Research Advisory Board; and Oak Ridge National Laboratory (ORNL). 1976. Ecology and Analysis of Trace Contaminants, Progress Report, October 1974-December 1975. ORNL/NSF/EATC-22. Oak Ridge, Tennessee. 200 pp.

[e]PCM = Poplar Creek Mile.

[f]This study, based on data for July 1975.

It is highly probable that significant discharges of Hg were occurring from New Hope Pond into the East Fork of Poplar Creek prior to the time in 1971 when Hg-contaminated sediments in the pond were pumped to a holding basin on Chestnut Ridge. Because of the affinity of Hg for organic ligands, it is likely that most of the mercury discharged from New Hope Pond was associated with suspended particulate materials (i.e., algae). Concentrations of Hg associated with suspended particulate matter in New Hope Pond are not measured in the nonradiological monitoring program at Y-12, and monitoring data for the New Hope Pond discharge in 1971, 1972, and 1973 (UCCND 1972, UCCND 1973, UCCND 1974) do not indicate elevated levels of dissolved Hg in pond water. However, reported monitoring data for dissolved Hg are based on water samples which were composited over a week, filtered and then acidified with HNO_3. In view of the known instability of dissolved mercury in water samples, due to a combination of bacterial uptake and adsorption onto container walls, high volatility, sorption of Hg by filter material and formation of stable complexes with other organic materials in suspension (Avotins and Jenne 1975, Jenne and Avotins 1975), it is likely that reported concentrations of dissolved Hg (i.e., that Hg which passes through a membrane filter with an ~ 0.4-μ pore size) in the New Hope Pond effluent (and in other water samples collected and processed in a similar manner) are in error on the low side. Thus, considering the fact that total Hg in the New Hope Pond effluent is not monitored, and measured concentrations of dissolved mercury in composite water samples probably underestimate actual concentrations released during

the sampling period, it is not possible to state with certainty that elevated levels of Hg in sediments and fish in EFPC-PC are not due, in part, to recent discharges from New Hope Pond.[5]

Another potential source of the elevated mercury levels in the Poplar Creek-Clinch River drainage is the City of Oak Ridge's sewage treatment plant located on the East Fork of Poplar Creek. Mercury contamination of sediments collected near municipal sewage treatment plants has been reported by several investigators (e.g., Cranston and Buckley 1972, Oliver 1973, Van Loon 1974). The increased mercury levels in sediments have been attributed to direct discharges of mercury in the liquid effluents from these sewage treatment facilities. In some cases, however, mercury contamination in aquatic sediments also has resulted from erosion and runoff of sewage sludge which has been shown to contain elevated mercury levels (Van Loon 1974).

The fact that elevated Hg concentrations have been found in the deeper strata of core samples from the East Fork of Poplar Creek above and below the sewage treatment plant and from Poplar Creek (ERDA 1975a, Reece 1974), indicates that sediments in this drainage contaminated from previous releases of mercury have not been entirely scoured and transported downstream. Studies in Sweden and the United States have shown that mercury in contaminated lake sediments remains biologically available to aquatic food chains 15 to 25 years after emissions were stopped (Wallace et al. 1971, Jernelöv et al. 1975). Estimates of the "ecological half-time" (i.e., the time required for concentrations in aquatic food chains to decrease by one-half) for Hg in contaminated Swedish lakes after all discharges were

stopped have ranged from 10 to 100 years (Jernelöv et al. 1975).
Biological half-lives for mercury in fish alone have been estimated
to range from 1 to 3 years (e.g., Burrows and Krenkel 1973, McKim
et al. 1976). It is possible, therefore, that the elevated levels of
Hg in sediments and fish in the East Fork of Poplar Creek, Poplar
Creek, and the Clinch River are a result of previous discharges which
have been retained in the sediments of these environs and which are
still biologically available to aquatic food chains.

It is clear that the EFPC and PC from PCM 0 upstream to the con-
fluence with EFPC are contaminated with mercury, based in comparisons
of mercury levels in sediments and fish from both control areas and from
reported concentrations in uncontaminated reservoirs and rivers in the
area. It is also apparent that mercury discharged into the PC drainage
has been transported into the Clinch River downstream of PC, although
the extent and distribution of the mercury contamination in the CR
are not known. The available data at the time indicates that signifi-
cant Hg contamination of aquatic food chains, in terms of Hg levels
that equal or exceed the FDA's proposed limit of 0.5 ppm Σ Hg in edible
portions of fish, is limited to largemouth bass in PC and in the CR
at the mouth of PC. Although the data are limited, elevated Hg concen-
trations in fish from the East Fork of Poplar Creek, Poplar Creek,
and the Clinch River were measured in 1970 (Sanders 1970, Bolton and
Whitson 1971); the significance of the observed levels apparently was
not fully appreciated at that time. Monitoring of Hg levels in fish
or other biota from these environs, unfortunately, was not done, so
the trend in concentrations over the past six years cannot be determined.

Recommendations

(1) Studies should be initiated to identify the source(s) and speciation of mercury in the East Fork of Poplar Creek, Bear Creek, Poplar Creek, and the Clinch River by determining the spatial distribution of mercury associated with sediments of selected size fractions and organic content in these environs. It is suggested that mercury associated with surface sediments be mapped in detail in New Hope Pond, the East Fork of Poplar Creek, Bear Creek, and Poplar Creek, above and below the confluence with the East Fork of Poplar Creek, and the Clinch River above and below the mouth of Poplar Creek.

(2) Additional sampling of aquatic food chains (i.e., benthic invertebrates, fish) in the East Fork of Poplar Creek, Poplar Creek, Bear Creek, and the Clinch River should be conducted in order to quantify the baseline levels of Hg contamination in biota in 1977 for comparison with future monitoring data. Fish sampling should be initiated in April in order to collect migrating species such as sauger and white bass which spawn in the Poplar Creek drainage. The sampling should be designed to collect all life history stages (i.e., eggs, fry, fingerlings, and adults of all age groups and sexes) of the species of interest. Analysis for Hg content of components of aquatic food chains in these contaminated environs and in control (uncontaminated areas should be initiated as soon as possible and conducted on a regular basis (e.g., minimum of annually).

(3) The Quality Control Program for environmental monitoring of mercury (and other heavy metals) should be upgraded. Methods of collection, preparation, and analysis of environmental samples

for Hg should be standardized so that data collected in the various monitoring programs at X-10, Y-12, and K-25 are comparable. For sediment samples, it is recommended that the procedure for sampling surface sediments be carefully defined and standardized and the < 63-μ size fraction (i.e., sediment particles that pass through a U.S. Standard 250-mesh sieve or a 230 Tyler Standard Sieve) be analyzed. Total mercury in the two size fractions (i.e., > 63 and < 63 μ) and in bulk sediments should be analyzed in select samples in order to check recoveries and to determine the distribution of mercury as a function of particle size. Sediment samples should be wet sieved (using filtered water from the site where the sample was collected) as soon as possible after collection (and prior to freezing), dried at < 60°C, and quantitatively analyzed for Hg using flameless AAS. The dissolution method of sediment samples should follow the procedure recommended by the Environmental Protection Agency (see "Mercury in Sediments" on p. 134 of USEPA 1974). Quality control samples, including "blind" reference standards for water, sediments, and biological materials, should be exchanged by all monitoring laboratories on a regular basis.

(4) Procedures for monitoring mercury in liquid effluents and surface waters should be modified so that both total and particulate mercury are measured. Dissolved mercury can then be determined by the difference. In addition, analysis should be done on grab samples collected under various flow regimes rather than on composite samples held over a week. Water samples should be collected in glass containers and acidified immediately after collection with concentrated Ultrex nitric acid. Holding times prior to analysis for mercury

samples should be kept to a minimum, preferably < 12 hr. For particulate Hg, aliquots should be filtered immediately on collection through a membrane filter (Nuclepore membrane filters are recommended because of their uniform pore size, low Hg background, and low sorption of Hg by filter material) with a 0.4-μ pore size before adding the acid. For total Hg, the filtration is omitted, and the sample should be collected directly in a pre-acidified, baked glass volumetric container.

(5) Because of the agricultural utilization of the East Fork of Poplar Creek and its floodplain and the potential for deposition for Hg-contaminated sediments on the floodplain during high discharges, it is recommended that soils and vegetation along the East Fork of Poplar Creek be sampled and analyzed for Σ Hg.

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Footnotes

[1]The FDA's proposed action level for mercury in fish and shellfish was
first established in 1969 and reaffirmed in 1974 after several reviews
of the proposed limit. The 0.5 limit of Σ Hg is based on the assump-
tions that all mercury in fish and shellfish is methylmercury (MeHg)
and the per capita consumption of fish is 60 g/day. Consuming this
amount of fish containing 0.5 ppm MeHg will result in a daily inges-
tion rate of 0.03 mg MeHg, a level which still maintains a 10-fold
margin of safety over the level of mercury intake known to be toxic
to humans. While the 0.5 ppm limit for Σ Hg in fish is still considered
a proposed action level, it is being applied and enforced by the FDA
as a compliance standard for fish and shellfish involved in interstate
commerce (Curtis Coker, FDA, Bureau of Foods, Division of Regulatory
Guidance, Washington, DC - personal communication). For fish not
involved in interstate commerce (such as commercial fish and fish
products which are caught, processed, and consumed within the same
state or sport fish), the agency has no legal jurisdiction to seek or
enforce compliance with the 0.5 ppm action level. In such cases, the
FDA can only work with agencies of the state governments responsible
for food safety and water pollution control, encouraging them to take
counter measures necessary to protect the public health. According to
Curtis Coker from the FDA, some states feel the 0.5 ppm proposed action
level for Hg in fish and shellfish is too low and in certain cases,
have chosen not to take counter measures when this level was exceeded.

[2]A national survey of fish consumption patterns conducted by NOAA's
National Marine Fisheries Service in 1969-1970 (Finch 1973), showed

that only 1.8% of the U.S. population consumed an average of more than 60 g of fish and shellfish per day, and 1% of the participants surveyed consumed an average of 77 g per day. The average per capita consumption of fish and shellfish in the U.S. in 1969-1970 was estimated to be approximately 5 g/day. In 1975, the average per capita consumption of fish in the U.S. was estimated to be in excess of 7300 g/year (or equivalent to 20 g/day), 22% of which was conservatively estimated to come from sport fishing (Stroud 1977).

[3]Since May 1970, TVA systematically sampled fish from Pickwick Reservoir as well as from several other reservoirs and rivers in the TVA system. Contamination above the 0.5 ppm proposed action level was found only in the North Fork of the Holston River, in Pickwick Reservoir, and in Kentucky Lake, the reservoir immediately downstream from Pickwick. Average Σ Hg concentrations in fish in both reservoirs were found to exhibit a marked seasonal fluctuation with maximum concentrations in the summer and minimum concentrations in the fall and winter (Simpson et al. 1974). The decision to close Pickwick Reservoir to commercial fishing and to warn sport fishermen of the Hg-contaminated fish was made on the basis of the high Σ Hg levels obtained during the summer of 1970. However, fish sampling in Kentucky Lake was begun during late fall and early winter, near the seasonal minimum, as a result of which the lake was not closed. The Tennessee River area of Kentucky below Kentucky Dam was closed to commercial fishing in 1971 because of Hg contamination (Simpson et al. 1974). The public was also warned of the mercury contamination in fish from the North Fork of the Holston River below Saltville, Virginia (Wallace et al. 1971).

[4]Reanalysis of the two samples containing 300 ppm Σ Hg has confirmed the high values (Mike Mitchell, ORGDP, personal communication). Although the source of the mercury in these extraordinarily high concentrations is unknown, detailed mapping of Hg in sediments might help in defining the source.

[5]Since the upper portion of EFPC (i.e., the section from immediately below New Hope Pond to where EFPC crosses Gamble Valley Road) is a shallow turbulent stream, one would expect the fine-grained sediments (silt, clay) contaminated from historic releases of Hg from New Hope Pond to have been scoured out of this section and transported downstream. If this were the case and if there were no recent Hg discharges from New Hope Pond, one would also expect the recently deposited surface sediments in this upper portion of EFPC to contain Hg concentrations similar to those in uncontaminated streams in this area. Detailed sampling of recent fine-grained surface sediments in this portion of EFPC should confirm whether there have been recent discharges of Hg from New Hope Pond. Sediment samples should be collected by locating areas of "recent" deposition of fine-grained sediment (silt, clay). These areas are usually behind anchored debris and in slow-moving pools and back water areas. The top 1-2 cm layer of sediment should be scraped off and placed in a sample container. If analysis is delayed, samples should be frozen. Since freezing will change the texture of sediments, samples should be sonified to disperse aggregates. Biasing field collections to sample only fine-grained sediments is one approach to achieving textural uniformity of sediment samples. This can be determined by feeling the sediments to see if there are coarse (sand size or larger) particulates

in the samples. If the sediment samples are not of a uniform, fine-grained texture, mechanical sieving through a 63 μ mesh opening will be necessary.

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